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Bentonite Borehole Plug Flow Testing with Five Water Types

Prepared by M.V. Gaudette, J.J.K. Daemen

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Prepared for
U.S. Nuclear Regulatory Commission

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Bentonite Borehole Plug Flow Testing with Five Water Types

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ABSTRACT

The hydraulic conductivity has been determined of plugs constructed with commercial precompressed bentonite pellets. Bentonite has been hydrated and tested with waters of five different chemical compositions, including one groundwater (Ogallala aquifer, Texas). The groundwater contained a significant amount of solids; waters prepared in the laboratory did not. Prepared waters used for testing included distilled water, a high (1000 ppm) and a low (45 ppm) calcium solution, and a 39 ppm sodium water. Uncompacted plugs were constructed by dropping bentonite tablets into waterfilled cylinders, or by mixing powdered bentonite with preselected water volumes in order to obtain controlled initial water contents. The hydraulic conductivity of all plugs tested with all waters would result in a classification of practically impervious, by conventional soil mechanics standards. Variations of several orders of magnitude of the hydraulic conductivity are observed. Clay plugs constructed from bentonite tablets hydrated with unfiltered Ogallala groundwater exhibited reduced swelling and lower hydraulic conductivity than similar plugs constructed from tablets and distilled water. The differences in observed conductivities may be governed by factors including swelling characteristics and permeant colloidal matter clogging plug pore spaces. Constant pressure injection and transient pulse testing methods are used to determine hydraulic conductivity. Especially in constant pressure injection tests, outflow volumes may require adjustment to account for consolidation drainage. Attempts made here at such corrections have not been successful. Consolidation testing requires extremely long time periods to approach a constant limit. It appears probable that the flow tests have not been pursued for a sufficient time to assure complete determination of water chemistry effects on hydraulic conductivity. Nevertheless, all indications are that simple conventional bentonite tablet borehole plug construction can produce a borehole plug with low hydraulic conductivity. It needs to be recognized that such plugs may very well contain relatively large voids and inhomogeneities. As a result, the hydraulic conductivity is markedly higher than values typically quoted in the literature for bentonite, most of which appear to be derived for carefully prepared, uniform, compacted and very small bentonite samples. It is clear that values used in performance assessments for borehole and shaft seals need to be based on a comprehensive determination of in-situ conditions, or need to be based on conservative limiting conditions.

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SYMBOLS AND ACRONYMS

Roman Alphabet

A:	flow sample cross-sectional area
C_o:	bulk solution ion concentration
C:	ion concentration
C_c:	coefficient of consolidation
CEC:	cation exchange capacity
d:	representative porous medium length
d_p:	particle diameter
d_M:	pore diameter of a medium
d_{50}:	mean grain diameter
D:	dielectric constant
DDL:	diffuse double layer
e:	void ratio
e:	electronic charge
ESP:	exchangeable sodium percentage
ESR:	exchangeable sodium ratio
F:	force
GTP:	Generic Technical Position
h:	hydraulic head
h_d,h_u:	downstream and upstream hydraulic heads in transient pulse testing
H:	upstream head applied in transient pulse test
k:	Boltzman constant
K:	hydraulic conductivity
K_g:	Gapon reaction coefficient
K_k:	reaction coefficient
ℓ:	transient pulse test sample length
L:	flow sample length
M_v:	coefficient of volume change
n:	porosity
NRC:	U.S. Nuclear Regulatory Commission
NTU:	nephelometric turbidity units
P:	pore pressure

q: specific discharge

q, q': electrical charge

Q: flow rate

r: charge separation

R_e: Reynolds number

SAR: sodium adsorption ratio

S_d: compressive storage

S_s: compressive storage

t: time

T: absolute temperature

V_A: attractive energy

V_R: repulsive energy

V: volume

V_v: void volume

x: distance (position) along a sample

X: exchangeable cation form

Z: ion valence

Greek Alphabet

α: dimensionless time (in transient pulse analysis)

α: medium compressibility (in specific storage calculation)

β: fluid compressibility (in specific storage calculation)

β: ratio of sample compressive storage to upstream reservoir compressive storage in transient pulse test (Eqtn. 4.12)

ψ: electrical potential

γ: ratio of downstream to upstream reservoir compressive storage in transient pulse test (Eqtn. 4.13)

ν: kinematic viscosity

ρ: density

σ_e: effective stress

EXECUTIVE SUMMARY

The hydraulic conductivity has been determined of plugs constructed with commercial precompressed bentonite pellets. Bentonite has been hydrated and tested with waters of five different chemical compositions, including one groundwater (Ogallala aquifer, Texas). The groundwater contained a significant amount of solids; waters prepared in the laboratory did not. Prepared waters used for testing included distilled water, a high (1000 ppm) and a low (45 ppm) calcium solution, and a 39 ppm sodium water.

Uncompacted plugs were constructed by dropping bentonite tablets into waterfilled cylinders, or by mixing powdered bentonite with preselected water volumes in order to obtain controlled initial water contents. The hydraulic conductivity of all plugs tested with all waters would result in a classification of practically impervious, by conventional soil mechanics standards. Variations of several orders of magnitude of the hydraulic conductivity are observed.

Clay plugs constructed from bentonite tablets hydrated with unfiltered Ogallala groundwater exhibited reduced swelling and lower hydraulic conductivity than similar plugs constructed from tablets and distilled water. The differences in observed conductivities may be governed by factors including swelling characteristics and permeant colloidal matter clogging plug pore spaces.

Constant pressure injection and transient pulse testing methods are used to determine hydraulic conductivity. Especially in constant pressure injection tests, outflow volumes may require adjustment to account for consolidation drainage. Attempts made here at such corrections have not been successful. Consolidation testing requires extremely long time periods to approach a constant limit.

It appears probable that the flow tests have not been pursued for a sufficient time to assure complete determination of water chemistry effects on hydraulic conductivity. Nevertheless, all indications are that simple conventional bentonite tablet borehole plug construction can produce a borehole plug with low hydraulic conductivity. It needs to be recognized that such plugs tend to contain relatively large voids and inhomogeneities. As a result, the hydraulic conductivity is markedly higher than values typically quoted in the literature for bentonite, most of which appear to be derived for carefully prepared, uniform, compacted and very small bentonite samples. It is clear that values used in performance assessments for borehole and shaft seals need to be based on a comprehensive determination of in-situ conditions, or need to be based on conservative limiting conditions. It is not likely that measurements on small samples prepared with unusual care can produce numerical values that represent realistic bounding conditions corresponding to large-volume in-situ emplacement situations.

The introductory chapter of this report briefly summarizes the regulatory context of rock mass sealing requirements, quotes commentaries and viewpoints on rock mass sealing from the literature on HLW disposal, outlines the organization of this report, and lists related reports issued under this contract.

The primary seal component studied here, bentonite, is described in Chapter Two. This chapter includes a brief summary of essential behavioral aspects of bentonite that are likely to influence its sealing performance. Included is a description of the bentonite used in the experiments reported on. Also included are selected references that point out the complexity of the material, the sensitivity of its sealing performance (e.g. hydraulic conductivity, strength) to numerous installation and environmental variables, as well as to its intrinsic properties and characteristics.

Chapter Three presents a similar introduction to properties and characteristics of the permeant, water, that are likely to influence the sealing performance of bentonite. A brief discussion of chemical and physical characteristics is included. Some of these permeant characteristics can have a major impact on measured hydraulic conductivities, and it appears quite essential that their effects be fully accounted for in any assignment of numerical values to bentonite sealing performance. Also included in this chapter is a section in which the significant characteristics and properties of the waters used in these experiments are described.

Background on hydraulic conductivity testing of bentonite is given in Chapter Four. This includes a brief review of conventional testing, but emphasizes strongly transient pulse testing because of the potential this method has for accelerating the otherwise exceedingly time-consuming testing. Also included is a discussion of consolidation, as consolidation effects are likely to affect the tests, and, in that case, need to be accounted for.

Chapter Five presents the plug installation and flow testing, including test equipment and test procedures. Pipe flow tests are described, in which bentonite is installed in plastic tubing. Experimental results are given in Chapter Six. Conclusions are drawn in Chapter Seven, which also identifies remaining uncertainties and information needs, from which recommendations are derived for follow-up research.

Appendix A provides comprehensive hydraulic conductivity curves, Appendix B gives mass balance plots, and Appendix C consolidation plots. Appendix D gives theoretical type curves as well as the actual hydraulic head decay curves obtained from transient pulse testing of bentonite plugs.

CHAPTER ONE

INTRODUCTION

1.1 Objective

The primary objective of this study is to determine how the hydraulic conductivity of bentonite is affected by water permeants having varying compositions. A groundwater, distilled water, and three synthetic waters, are used in evaluating the influence of water permeants on the hydraulic conductivity of bentonite. The groundwater used in this study is from the Ogallala aquifer in northern Texas, near Lubbock. The synthetic waters are made by dissolving the appropriate amount of sodium chloride and calcium chloride in distilled water to obtain two distinct waters having sodium and calcium concentrations similar to those present in the groundwater. Additionally, a relatively high concentration calcium water was made in a similar fashion.

Hydraulic conductivity testing is accomplished using two methods, transient pulse testing and more conventional constant pressure injection testing. A secondary objective of this study is to attempt to evaluate the applicability of transient pulse testing in determining the hydraulic properties of bentonite. Hydraulic conductivity values for bentonite obtained from the literature and from constant pressure injection testing are compared with values obtained from transient pulse testing. Calculated specific storage values for bentonite are compared with those obtained by transient pulse testing. If a reasonable correlation could be demonstrated to exist between expected and actual values, it would indicate transient pulse testing to be a relatively quick, reliable method for determining the hydraulic properties of bentonite.

The fundamental objective of this "Rock Mass Sealing" research project is to assess experimentally the performance of existing products and methods for sealing rock masses, in the current phase of the project to conduct an experimental evaluation of borehole plug performance. This work is aimed at determining the feasibility of sealing boreholes intersecting a repository rock mass to a level where it can reasonably be assured that the plugged boreholes will not become preferential radionuclide migration paths. This project studies experimentally the likelihood of preventing such migrations by sufficiently reducing the hydraulic conductivity of the plugged borehole (including the plug-rock interface). The work performed provides direct input to an assessment as to whether or not 10 CFR 60.134, Design of seals for shafts and boreholes (U.S. Nuclear Regulatory Commission, 1983, and, as amended, 1985, 1987) is likely to be satisfied by any particular proposed sealing methods. Some aspects of this study have broader implications, e.g. directly for 10 CRF.133 (d), and indirectly for 10 CFR.133 (a)(1), (U.S. Nuclear Regulatory Commission 1983a, and as amended, 1985, 1987), (h).

The study is conducted to establish a factual data basis on borehole sealing performance, to assess the applicability of seal testing procedures, and, especially for this report, to identify some site characteristics, particularly ground water chemistry, that may need to be identified in order to assess sealing performance and make sealing performance predictions. The latter aspect is of particular significance within the context of repository performance assessments and component performance allocation. Although some types of borehole sealing have been performed for many years, relatively little testing and sealing performance verification has been reported on.

Concern about boreholes and their potential influence on the isolation performance of the rock mass surrounding repositories has been expressed in a number of basic reviews on undergrcund HLW (High level radioactive waste) disposal (e.g. Atomic Energy of Canada Limited, 1978, p. 72; Bredehoeft et al., 1978, p. 8; Committee on Radioactive Waste Management, 1978, pp. 5,10; Heineman et al., 1978, p. 4; U.S. Department of Energy, 1979, p. 3.1.328; Arnett et al., 1980, p. 139; Barbreau et al., 1980, p. 528; Burkholder, 1980, p. 15; Irish, 1980, p. 42; OECD, 1980, Foreword; Pedersen and Lindstrom-Jensen, 1980, p. 195; U.S. Department of Energy, 1982, p. 29; Deju, 1983, p. 4; Kocher et al., 1983, p. 54; National Research Council, 1983, p. 8-9, 21, 63; U.S. Department of Energy, 1983, p. 25; Pigford, 1983, p. 10).

Some years ago it was widely considered desirable, if not necessary, that sealing be performed such that the plug be at least as impermeable as the rock it replaces (e.g. Carlsson, 1982; Fernandez and Freshley, 1984, p. 43). Such a requirement was incorporated in the Proposed Rule 10 CFR Part 60 (U.S. Nuclear Regulatory Commission, 1981, §60.133), but has been relaxed in the Final Rule (U.S. Nuclear Regulatory Commission, 1983a, § 134(a)), for reasons discussed at some length in Staff Analysis of Public Comments (U.S. Nuclear Regulatory Commssion, 1983b, pp. 72-72, 422-430).

The need for borehole plugging, and particularly for very high performance (e.g. very low hydraulic conductivity), is no longer universally accepted, nor obvious, and certainly might be a somewhat site dependent requirement, as shown by consequence assessments (e.g. Pedersen and Lindstrom-Jensen, 1980, p. 195; Klingsberg and Duguid, 1980, p. 43; Intera Environmental Consultants, Inc., 1981). These authors do recognize that borehole seals will provide "... an important redundant barrier ..." or "... will satisfy the concept of multiple barriers ...". A panel of experts convened by the Commission of the European Communities and the OECD Nuclear Energy Agency simultaneously considers backfilling and sealing (OECD, 1984, Section III.4) and, after stressing the host rock specificity of backfill and sealing functions, states that "they (i.e. seals) would be designed so as not to present any preferential flow paths ... there is confidence that they (i.e. the functional requirements) can be met by a number of different materials." That the controversy about sealing requirements is far from resolved is particularly well illustrated by the recently published disagreements among the ONWI Exploratory Shaft Peer Review Group (Kalia, 1986, p. 14). Site specific analyses are most likely to provide information needed to finalize specific performance and design

requirements (e.g. Stormont, 1984; Freshley et al., 1985; Seitz et al., 1987).

General guidelines for the separation of radioactive waste from the physical environment, and in particular for the acceptable radionuclide releases following repository closure, have been finalized by EPA (U.S. Environmental Protection Agency, 1986; 40 CRF 191). Detailed implementation of the requirements is governed by 10 CFR 60 (U.S. Nuclear Regulatory Commission, 1983a, 1985). The research performed as part of this ongoing contract addresses specifically some of the remaining uncertainties associated with the sealing requirements in 10 CFR 60, including §60.51,(a)(4); §60.102,b(2),e(1),(2); §60.113; §60.133,(h), §60.142,(c), but particularly §60.134, Design of seals for shafts and boreholes.

Further guidance on implementation of NRC Rule 10 CFR 60 with respect to borehole and shaft sealing is provided in a Generic Technical Position (GTP) (U.S. Nuclear Regulatory Commission, 1986). The GTP identifies the information needs to be satisfied before construction authorization can be granted. It also states the need for including borehole and shaft seals in performance analyses. The work reported on here is in direct support of providing NRC with independent information and assessment tools for reviewing the corresponding parts of an eventual license application. Information of this type also is an integral part of the necessary input for repository performance assessments, in this case specifically with regard to post-closure performance, especially the performance of bentonitic borehole and shaft seals with regard to isolation. Finally, the data should be of use for model validation, particularly models that claim to be able to represent the flow of water through bentonite.

The present report addresses several regulatory requirements and potential concerns with regard to sealing of boreholes and shafts with bentonite. These are amplified in Chapter Seven, Conclusions, and only highlighted here.

Rule 10 CFR Part 60 142 requires in-situ seal testing. This report, as well as several related ones, points out the extreme difficulty in testing bentonite. Of particular concern is the need for quite detailed determination and simulation of in-situ environmental conditions. These requirements already have been specified in the Borehole and Shaft Sealing Generic Technical Position (U.S. Nuclear Regulatory Commission, 1986), e.g. Section 4.2, Seal Material Characteristics. The present report points out that hydraulic conductivity of a bentonite seal material can be defined only under very specific environmental conditions. Moreover, the sensitivity of the results to installation, groundwater composition, confinement, applied stress and hydraulic gradients, to name only factors investigated here, is such that extreme care will need to be exercised in identifying likely performance of seals of this type. Consideration may be given to evaluate the feasibility of requiring or assigning a performance (e.g. hydraulic conductivity) with a considerable safety or uncertainty margin. If such performance is acceptable within the context of overall repository performance, it may alleviate greatly the complexity of performance demonstrations. All indications are that the

latter will be exceedingly difficult if an absolute need exists for assessing in detail all parameters that may affect bentonitic seal performance, particularly over a prolonged period of time, i.e. a time sufficiently long to imply the occurrence of significant changes in environmental parameters that may affect bentonite sealing.

1.2 Scope and Limitations

An experimental investigation was conducted to examine the influence of water composition on the sealing performance of bentonite borehole plugs. The plugs were constructed with commercial well-sealing grade bentonite pellets, mixed with various water types, and flow tested with five different water types. Test durations have ranged up to six months. Most experiments have been conducted under relatively high gradients. Most plug installations were kept extremely simple, typically with pellets and minimal other preparation. All experiments were conducted at room temperature.

1.3 Organization

This introductory chapter briefly summarizes the regulatory context of rock mass sealing requirements, quotes commentaries and viewpoints on rock mass sealing from the literature on HLW disposal, outlines the organization of this report, and lists related reports issued under this contract.

The primary seal component studied here, bentonite, is described in Chapter Two. This chapter includes a brief summary of essential behavioral aspects of bentonite that are likely to influence its sealing performance. Included is a description of the bentonite used in the experiments reported on here. Also included are selected references that point out the complexity of the material, the sensitivity of its sealing performance (e.g. hydraulic conductivity, strength) to numerous installation and environmental variables, as well as to its intrinsic properties and characteristics.

Chapter Three presents a similar introduction to properties and characteristics of the permeant, water, that are likely to influence the sealing performance of bentonite. A brief discussion of chemical and physical characteristics is included. Some of these permeant characteristics can have a major impact on measured hydraulic conductivities, and it appears quite essential that their effects be fully accounted for in any assignment of numerical values to bentonite sealing performance. Also included in this chapter is a section in which the significant corresponding characteristics and properties of the waters used in these experiments are described.

Background on hydraulic conductivity testing of bentonite is given in Chapter Four. This includes a brief review of conventional testing, but emphasizes strongly transient pulse testing because of the potential this method has for accelerating the otherwise exceedingly time-consuming testing. Also included is a discussion of consolidation, as consolidation effects are likely to affect the tests, and, in that case, need to be accounted for.

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Appendix A provides comprehensive hydraulic conductivity curves, Appendix B mass balance plots, and Appendix C consolidation plots. Appendix D gives theoretical type curves as well as the actual hydraulic head decay curves obtained from transient pulse testing of bentonite plugs.

1.4 Rock Mass Sealing Contract No. NRC-04-78-271 - Reports Issued

This Technical Report is the latest in a series of reports issued for the subject contract. A complete list of reports issued (to be issued for Schaffer and Daemen, 1987) is given below, to facilitate a general overview of work performed to date and of the overall context of ongoing work.

The first four reports, as well as the seventh, are literature surveys.

The fifth report is primarily a description of planning, experimental design and some preliminary tests.

The topical report by Jeffrey (1980) gives a comprehensive theoretical (analytical) discussion of transverse plug-rock interaction, based on elastic and viscoelastic calculations. This is complemented by the axial interaction discussed in Stormont and Daemen (1983), a report which is primarily experimentally oriented, but includes extensive analytical discussions.

The topical reports by Mathis and Daemen (1982) and by Fuenkajorn and Daemen (1986) present a detailed experimental assessment of drilling damage in granites and in basalts.

Experimental flow studies under polyaxial stress conditions are described in Cobb and Daemen (1982), under radially symmetric external loading in South and Daemen (1986), and on unloaded samples in Akgun and Daemen (1986). Additional data on plug performance under stressed and unstressed conditions are included in virtually all other reports.

Schaffer and Daemen (1987) describe experiments on rock fracture grouting, emphasizing the considerable lack of detailed knowledge and need for further research in this area.

Kimbrell et al. (1987) investigate the field performance of cement and of bentonite plugs installed in boreholes in granites.

Williams and Daemen (1987) report flow tests on borehole plugs constructed of mixtures of crushed rock (basalt) and bentonite.

Sawyer and Daemen (1987) describe experimental work on bentonite borehole plugs constructed by various means of seven commercial well-sealing grade bentonites, including flow tests and strength tests.

All annual reports subsequent to (5) include a combination of experiments, results, conclusions, and plans for future work, similar to the present annual report.

Quarterly progress reports are not listed as all information contained therein also is included in the annual reports.

1. South, D.L., R.G. Jeffrey, L.W. Klejbuk, and J.J.K. Daemen, 1979, "Rock Mass Sealing - Annual Report, October 1, 1978 - September 30, 1979," prepared for the U.S. Nuclear Regulatory Commission, SAFER Division, for Contract NRC-04-78-271, by the Department of Mining and Geological Engineering, University of Arizona, Tucson.

2. Daemen, J.J.K., 1979, "Rock Mass Sealing (Research in Europe)," 48 pp., Foreign Travel Trip Report to the U.S. Nuclear Regulatory Commission, SAFER Division, for Contract NRC-04-78-271, by the Department of Mining and Geological Engineering, University of Arizona, Tucson.

3. South, D.L., 1979, "Well Cementing," 75 + vii pp., Topical Report to the U.S. Nuclear Regulatory Commission, SAFER Division, for Contract NRC-04-78-271, by the Department of Mining and Geological Engineering, University of Arizona, Tucson.

4. Sultan, H.A., "Chemical Grouting for Rock Mass Sealing - a Literature Review," 45 + iv pp., Topical Report to the U.S. Nuclear Regulatory Commission, SAFER Division, for Contract NRC-04-78-271, by the Department of Mining and Geological Engineering, University of Arizona, Tucson.

5. South, D.L., R.G. Jeffrey, S.L. Cobb, S.P. Mathis, and J.J.K. Daemen, 1979, "Rock Mass Sealing - Annual Report, October 1, 1979 - September 30, 1980," prepared for the U.S. Nuclear Regulatory Commission, SAFER Division, for Contract NRC-04-78-271, by the Department of Mining and Geological Engineering, University of Arizona, Tucson.

6. Jeffrey, R.G., 1980, "Shaft or Borehole Plug-Rock Mechanical Interaction," 145 + xi pp., Topical Report to the U.S. Nuclear Regulatory Commission, SAFER Division, for Contract NRC-04-78-271, by the Department of Mining and Geological Engineering, University of Arizona, Tucson.

7. South, D.L., 1980, "Borehole Sealing with Clay (Part A). Considerations in Clay Mineral Stability (Part B)," 50 + iv pp.; 25 + ii pp., Topical Report to the U.S. Nuclear Regulatory Commission, SAFER Division, for Contract NRC-04-78-271, by the Department of Mining and Geological Engineering, University of Arizona, Tucson.

8. Cobb, S.L., W.B. Greer, R.G. Jeffrey, S.P. Mathis, D.L. South, and J.J.K. Daemen, 1981, "Rock Mass Sealing - Annual Report, September 1, 1980 - May 31, 1981," prepared for the U.S. Nuclear Regulatory Commission, SAFER Division, for Contract NRC-04-78-271, by the Department of Mining and Geological Engineering, University of Arizona, Tucson.

9. South, D.L., W.B. Greer, N.I. Colburn, S.L. Cobb, B. Kousari, S.P. Mathis, R.G. Jeffrey, C.A. Wakely, and J.J.K. Daemen, 1982, "Rock Mass Sealing - Annual Report, June 1, 1981 - May 31, 1982," prepared for the U.S. Nuclear Regulatory Commission, SAFER Division, for Contract NRC-04-78-271, by the Department of Mining and Geological Engineering, University of Arizona, Tucson.

10. Mathis, S.P. and J.J.K. Daemen, 1982, "Borehole Wall Damage Induced by Drilling: An Assessment of Diamond and Percussion Drilling Effects," 171 + xii pp., Topical Report to the U.S. Nuclear Regulatory Commission, SAFER Division, for Contract NRC-04-78-271, by the Department of Mining and Geological Engineering, University of Arizona, Tucson.

11. Cobb, S.L. and J.J.K. Daemen, 1982, "Polyaxial Testing of Borehole Plug Performance," 180 + xi pp., Topical Report to the U.S. Nuclear Regulatory Commission, SAFER Division, for Contract NRC-04-78-271, by the Department of Mining and Geological Engineering, University of Arizona, Tucson.

12. Daemen, J.J.K., D.L. South, W.B. Greer, J.C. Stormont, S.A. Dischler, G.S. Adisoma, N.I. Colburn, K. Fuenkajorn, D.E. Miles, B. Kousari, and J. Bertuca, 1983, "Rock Mass Sealing - Annual Report, June 1, 1982 - May 31, 1983," NUREG/CR-3473, prepared for the U.S. Nuclear Regulatory Commission, Division of Health, Siting and Waste Management, for Contract NRC-04-78-271, by the Department of Mining and Geological Engineering, University of Arizona, Tucson.

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CHAPTER TWO

BENTONITE, A POTENTIAL REPOSITORY SEALANT

2.1 Introduction

An issue which must be addressed when assessing the performance of a geologic repository for high-level nuclear wastes is the sealing of penetrations of the host rock. The United States Department of Energy (DOE), the agency responsible for developing licensed, high-level nuclear waste repositories, has stated that "penetrations into the host-rock system shall be sealed as necessary to exclude water, retard radionuclide migration and prevent communication between aquifers to the extent necessary for adequate isolation based upon performance assessments of the system" (U.S. Department of Energy, 1982, p. 29).

The Department of Mining and Geological Engineering at the University of Arizona has been investigating the performance of existing rock mass sealing technology (Section 1.4). This includes laboratory and field testing of borehole seals, theoretical analysis and data interpretation. Sealing materials investigated include cement, bentonite and bentonite-crushed basalt mixtures. This study investigates the influence a groundwater permeant has on the sealing properties of bentonite.

2.2 Bentonite, and its Sealing Capabilities

Knight (1898) suggested the name bentonite for a peculiar clay-like material found in the Fort Benton formation in Wyoming. Hewett (1917) and Wherry (1917) first established the origin of this material as an alteration product of volcanic ash. Condra (1908) suggested this origin for a similar clay in northern Nebraska. The following definition was offered by Ross and Shannon in 1926: "Bentonite is a rock composed essentially of a crystalline clay-like mineral formed by the devitrification and the accompanying chemical alteration of a glassy igneous material, usually a tuff or volcanic ash ... The characteristic clay mineral has a micaceous habit and facile cleavage, high birefringence, and a texture inherited from the volcanic tuff or ash, and it is usually the mineral montmorillonite." The term bentonite has become well established for any clay which is composed dominantly of a smectite clay mineral, and whose properties are dictated by this clay mineral (Wright, 1968). Whereas within a geological or mineralogical context the term does not necessarily imply certain physical properties or behavior (e.g. Grim, 1953, p. 361; Grim, 1968, p. 566), in geotechnical engineering the term refers to a highly plastic colloidal swelling clay (e.g. Mitchell, 1976, p. 39; Sowers, 1979, p. 86; Gillott, 1968, p. 176).

Bentonite has been used extensively to prevent water seepage through earthen structures and irrigation canals. It is effective in waterproofing the outsides of basement walls. Bentonite has been

employed as a liner in sanitary landfills. The Department of Energy has been considering bentonite as a component in an engineered barrier to prevent or retard the migration of water and radionuclides into or out of a nuclear waste repository (U.S. Department of Energy, 1986a, Section 5.1.4; 1986b, Section 6.4.2.2.1). These applications depend upon the sealant capabilities of bentonite.

The sealing capability of bentonite is partially described by its coefficient of permeability or hydraulic conductivity. The coefficient of permeability can be expressed as gallons per day per square foot. The Water Resources Division of the U.S. Geological Survey has replaced the coefficient of permeability with a term called hydraulic conductivity. This parameter is expressed in consistent units of length per time. A medium has a hydraulic conductivity of unit length per unit time if it transmits in unit time a unit volume of fluid at a specific viscosity through a cross section of unit area measured at right angles to the direction of flow, under a hydraulic gradient of unit change in head through unit length of flow (Lohman, 1979, p.6).

The hydraulic conductivity of a porous medium is a function of both the medium and the fluid percolating through it. Variables affecting the hydraulic conductivity include physical, chemical and biological characteristics of the matrix and of the fluid.

Physical parameters which affect the hydraulic conductivity of a porous medium, and specifically, bentonite, include texture and structure. Fine-grained soils have lower hydraulic conductivity values than coarse-grained soils. A soil having a mixture of grain sizes generally has a lower hydraulic conductivity than a soil of uniform grain size because some mixed soil pores are blocked by interstitial grains.

Compaction or consolidation reduces porosity and conductivity by breaking the natural soil structure or fabric. Pore volumes are reduced, resulting in smaller hydraulic conductivities. Pore volumes are also reduced during swelling of expansive soils.

The moisture content of a soil affects hydraulic conductivity. Generally, hydraulic conductivity increases with increasing soil moisture content (Lambe and Whitman, 1969, p. 292). However, swelling is induced by adsorption of water between clay mineral sheets, resulting in decreased pore volume and decreased hydraulic conductivity (Quirk, 1968; Rowell et al., 1969; Frenkel and Rhoades, 1978). The type of exchangeable cation (Section 2.5) also affects swelling and hydraulic conductivity. Sodium ions can adsorb thick water layers, resulting in high swelling. Grim (1962, pp. 244-245) suggests that thick water layers adsorbed on the surface of sodium-saturated montmorillonite impede water flow by reduction of pore volumes and because of the physical interference of the movement of fluid water against the water surface of adsorbed water. The relationship between soil moisture content and hydraulic conductivity is very complex.

Hydraulic conductivity can be affected by suspended solids in the percolating fluid. The porous medium acts as a filter, trapping many

of these particles. Accumulation of particle aggregates within soil pores results in decreased hydraulic conductivity.

Hydraulic conductivity can inadvertently be affected by laboratory testing. Some testing methods require that water be injected into a sample while the resulting effluent water is collected and measured. Air entrapment in the soil pores during wetting decreases the conductivity. The first common source for air introduction into a soil sample results from sample preparation. A sample may be prepared by adding granular material or tablets to water and allowing for a specific swelling time before commencing testing. The second source for air to be introduced to the soil sample is from the permeant. Dissolved gases may come out of solution and lodge within the pore space of the medium. These effects can be minimized by degassing all fluids before using them. Degassing can be accomplished by a number of methods, including boiling, bubbling an inert gas such as helium or nitrogen through the water, or by applying a vacuum to the water. The effect of air entrapment may also be reduced by wetting the sample from the bottom, causing air to be displaced at the top. This method may not be useful when clay samples are prepared by hydrating dry tablets. Neuzil (1986), in his landmark review of flow in low-permeability environments, lists an extensive set of experimental difficulties, and refers in particular to Mitchell and Younger (1967) and to Olson and Daniel (1981) for more detailed discussions.

Chemical parameters which affect the hydraulic conductivity of a porous medium include pH, cation exchange capacity, and exchangeable sodium percentage. These parameters may indicate the type and amount of chemical components in the soil and how they might interact with permeants to change hydraulic conductivity of the soil.

Biological parameters which affect the hydraulic conductivity of a porous medium include the macro effects of organics on the soil. These usually increase the hydraulic conductivity due to root penetration, worm, insect, and decayed root holes. Microorganisms acting on soil organic matter can either increase or decrease hydraulic conductivity. Microbial action can produce stable soil aggregates, resulting in higher conductivity values by clogging the soil pores with microbial cells or by-products of microbial metabolism (Allison, 1947).

Physical and chemical parameters affecting the hydraulic conductivity of a porous medium will be addressed in subsequent chapters. The biological parameters which influence the hydraulic conductivity of a porous medium are beyond the scope of this study.

2.3 Structure of Bentonite

The properties of bentonites depend upon the fact that they are made up of smectite clay minerals. The properties of smectite minerals in turn depend upon chemical composition, atomic structure, and morphology.

Bentonite is a term used to describe any clay which is composed dominantly of a smectite clay mineral, and whose properties are

determined by this clay mineral (Wright, 1968). It is not uncommon for bentonites to consist of at least 90 percent smectite. The bentonite utilized in this study contains a significant quantity of montmorillonite, a mineral belonging in the dioctahedral subgroup of smectite minerals. Montmorillonite is described by the following general formula (Bohn et al., 1985, p. 118):

$$(Al_{2-x}Mg_x)Si_4O_{10}(OH)_2Na_x^+$$

Montmorillonite is classified as a clay mineral, a term used to describe minerals having sheet silicate structures of the phyllosilicates. Most important clay minerals, such as montmorillonite, have somewhat related silicate structures. An understanding of silicate structures is therefore a logical starting point to study the permeability characteristics of bentonite.

The crystal structure of silicates is primarily determined by three controlling factors, (1) the type of chemical bonds in the crystal, (2) the crystal radii or size of the ions making up the crystal, and (3) the amount of substitution of one element for another during crystallization (Bohn et al., 1985, pp. 106-110).

Oxygen is a predominant ion in silicates. It constitutes nearly 50% of the mass and over 90% of the volume of most silicate structures. Oxygen has an ionic radius much larger than most cations found in silicates, making it a major factor in silicate structures. Silicate structures are determined largely by the way oxygen ions pack together. Ions pack around a central ion having a charge opposite to their own. The number of ions which can be packed around a central ion depends on the ratio of the radii of the two ions and is called the coordination number of the central ion. The coordination number of the central ion determines the atomic lattice of minerals. For example, the Si^{4+} cation is in fourfold, or tetrahedral, coordination with oxygen. The Al^{3+} cation is generally found in sixfold, or octahedral, coordination with oxygen, but can be in tetrahedral coordination. Tetrahedral and octahedral coordination units are the basic atomic lattices of most layer silicates.

The tetrahedral structure consists of a central Si^{4+} cation surrounded by four O^{2-} anions, giving the ionic unit $(SiO_4)^{4-}$ (Figure 2.1). The octahedral structure consists of six oxygen or hydroxyl groups packed around a central cation such as aluminum or magnesium resulting in a eight sided structure (Figure 2.2).

The montmorillonite clay mineral is a 2:1 layer silicate, composed of an octahedral sheet sandwiched between two silicon tetrahedral sheets (Figure 2.3). The tetrahedral sheets are bonded to the octahedral sheet by a layer of shared oxygen atoms. The tips of the tetrahedron point toward the center of the unit cell. The unit layers stack parallel to each other, hence the name layer silicates (Grim, 1953, pp. 55-64).

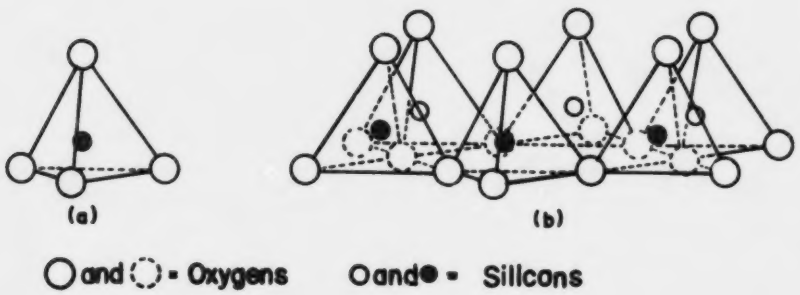

Figure 2.1 Schematic of the tetrahedral clay layer.

(a): Tetrahedron
(b): Tetrahedral layer (sheet) structure

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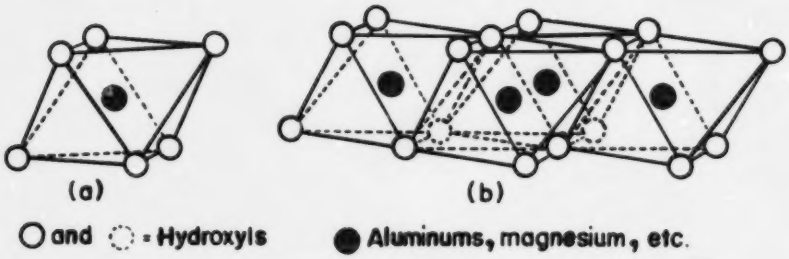

Figure 2.2 Schematic of the octahedral clay layer.

(a): Octahedron
(b): Octahedral layer (sheet) structure

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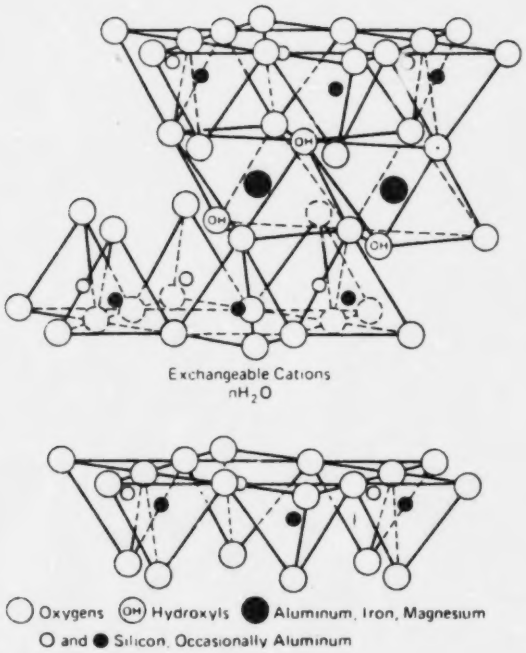

Figure 2.3 Schematic of the montmorillonite structure.

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The strength and type of interlayer bonding has a great influence on the properties of layer silicates. The type and strength of the interlayer bond is determined by the layer charge, defined as the charge per formula unit, when balanced by cations external to the unit layer. Layer charge can be explained by considering isomorphic substitution in silicate structures.

2.4 Isomorphic Substitution

Isomorphic substitution in silicate structures, the substitution of one element for another, is more the rule than the exception. Substitution takes place during crystallization and is not subject to substantial change afterwards. Ion size governs substitution. Substitution of ions having like charges may occur. Generally, substitution occurs between ions differing in size by less than 10 to 15% in ionic radii (Bohn et al., 1985, p. 110). Common isomorphic substitutions in silicates include Al^{3+} for Si^{4+} in the tetrahedral structure, and Mg^{2+}, Fe^{3+}, or Fe^{2+} for Al^{3+} in the octahedral structure. Substitution between ions of unlike charge results in excess charges within the crystal. This localized excess charge must be balanced by other cations or anions, either inside or outside the structural unit. Isomorphic substitution in silicates generally results in excess negative charge. To maintain electroneutrality, it is necessary to balance the excess negative charge with cations. The layer charge may therefore be described as excess charge per structural unit which is balanced by cations external to the unit layer.

Montmorillonites often exhibit imperfect isomorphic substitutions. Two important phenomena occur due to isomorphic substitution of trivalent cations in the crystal lattice with divalent cations, resulting in an excess negative charge on the unit cell. The first is cation exchange and the second is the formation of a diffuse double layer. Both greatly influence the permeability of bentonite and are discussed below.

2.5 Cation Exchange

Thompson (1850) and Way (1850) first studied cation exchange. They showed that passing an ammonium sulfate solution through soil resulted in a leachate consisting primarily of calcium sulfate. The predominant cation in solution changed from ammonium to calcium because of exchange with the soil (Bohn et al., 1979, p. 141). They found cation exchange to be stoichiometric, rapid, and reversible.

The cation exchange capacity of a clay mineral results from an overall negative charge on the mineral. The negative charge may be due to isomorphic substitution, broken chemical bonds, or substitution of hydrogen in hydroxyl ions (Mitchell, 1976, p. 130).

The cations exchanged in a cation exchange reaction are chemically equivalent and approximately stoichiometric. For this reason the cation exchange capacity, defined as the sum of all exchangeable cations present at a specified pH, varies only slightly with cation

species. Exchangeable cations are defined as cations which remain in solution but are associated with soil surfaces. Exchangeable cations and cation exchange capacity are normally expressed as moles of ion charge per kilogram of soil (moles + /kg).

Cation exchange reactions normally are virtually instantaneous. This may not be completely true for bentonite such as used in this study. The exchange reaction may require time to allow for ion diffusion to the mineral interlayers.

The ease with which one cation is replaced by another depends upon the charge, or valence, of the cation, and upon its hydrated radius. The lyotropic series describes the relative replacing power of certain cations (Bohn et al., 1985, p. 159; Mitchell, 1976, p. 131):

$$Li^{+} \approx Na^{+} > K^{+} \approx NH_4^{+} > Rb^{+} > C_s^{+} \approx Mg^{2+} > Ca^{2+} > Sr^{2+}\ Ba^{2+}$$
$$> La^{3+} \approx "H^{+}"(Al^{3+}) > Th^{4+} \qquad (2.1)$$

This series indicates that if a montmorillonite with adsorbed sodium ions comes into contact with an aqueous solution containing calcium ions, the calcium ions replace the sodium ions. The most important factor in determining the relative ease of cation replacement is the cation charge. Coulomb's law (1789) describes the attractive or repulsive forces between two particles such as the negatively charged clay mineral and an exchangeable cation. However, Coulomb's law breaks down when describing relative cation replacement between cations of the same valence. In order to describe such replacement, it is necessary to consider the hydrated radius of the cations involved.

Attraction of cations toward negatively charged soil colloids follows Coulomb's law (Bohn et al., 1979, p. 156; Mitchell, 1976, p. 113):

$$F = \frac{q\ q'}{D\ r^2} \qquad (2.2)$$

where F is the force of attraction or repulsion, q and q' are the electrical charges, D is the dielectric constant (78 for water at 25°C), and r is the charge separation. The strength of ion retention or repulsion increases with increasing ion charge q, with increasing colloid charge q', and with decreasing distance between the ion and colloid surface. Coulomb's law indicates that the most important factor in determining the relative strength of adsorption or desorption of a given ion is its valence. Trivalent cations will be held more strongly than divalent cations and divalent cations more strongly than monovalent ones.

Coulomb's law cannot be used to describe the preference of soil colloids for cations having the same valence. Such preference is related to the hydrated radius or energies of hydration of the ions.

Ions having a relatively small dehydrated radius have a higher charge density per unit volume and attract hydration water more strongly.

These hydrated cations have a larger radius than those of cations having a larger dehydrated radius. Cations having a large hydrated radius are held less tightly by clay minerals than cations having a smaller hydrated radius. Cations having a small hydrated radius can approach clay mineral surfaces much closer than cations with a larger hydrated radius. Within a given valence series, the degree of replaceability of an ion decreases as its dehydrated radius increases. It follows that a partially hydrated cation can approach clay mineral surfaces closer and is retained more tightly by the clay minerals.

Many equations have been used to describe cation exchange processes. Each has advantages and disadvantages. The use of one equation over another seems subjective and may be based as much on the location at which a given investigator was trained as on any other factor.

Cation exchange equations invoke assumptions which may not always be valid. These include: (1) separate cation or anion exchange is frequently considered, but their simultaneous presence is rarely acknowledged; (2) the cation or anion exchanger has a constant exchange capacity; (3) simple stoichiometry; and (4) complete reversibility.

The most general type of cation exchange reaction is a mass action equation of the form (Bohn et al., 1979, p. 154):

$$CaX + 2Na^{+} \leftrightarrows 2NaX + Ca^{2+} \qquad (2.3)$$

where X denotes the exchangeable form of the cation.

Solving for the reaction coefficient gives

$$K_k = \frac{(NaX)^2 (Ca^{2+})}{(CaX)(Na^{+})^2} \qquad (2.4)$$

Parentheses denote activities of soluble or exchangeable cations. Equation 2.4 can be rearranged to

$$\frac{(NaX)^2}{(CaX)} = K_k \frac{(Na^{+})^2}{(Ca^{2+})} \qquad (2.5)$$

which is commonly known as a Kerr-type exchange equation. It is valid for narrow concentration ranges. Kerr (1928) used ion concentrations rather than cation activity because of the difficulty in measuring or calculating activities precisely. Activities of ions in solution can be estimated from the Debye-Hueckel (1923) equation (Bohn et al., 1979, p. 154).

The Gapon (1933) equation used concentrations rather than activities of soluble ions and chemically equivalent quantities for both the colloid exchange sites and exchangeable cations:

$$(Ca)_{1/2}X + Na^{+} \rightleftharpoons NaX + 1/2Ca^{2+} \qquad (2.6)$$

The U.S. Salinity Laboratory (1954, p. 26) (see also Bohn et al., 1985, Section 9.2.4; Bohn et al., 1979, Section 8.2.4; Mitchell, 1976, p. 133) substituted the sum of calcium and magnesium for the exchangeable and bulk-solution calcium concentrations in the Gapon equation. This gives

$$\frac{[NaX]}{[CaX + MgX]} = K_g \frac{[Na^{+}]}{[Ca^{2+} + Mg^{2+}]^{1/2}} \qquad (2.7)$$

The left side of Equation 2.7 was termed the exchangeable sodium ratio (ESR), while the concentration term on the right side was named the sodium adsorption ratio (SAR), where

$$SAR = \frac{[Na^{+}]}{[Ca^{2+} + Mg^{2+}]^{1/2}} \qquad (2.8)$$

The concentration units are moles or mmoles per liter. When the concentration units are mmoles of charge per liter, equation 2.9 may be written as

$$SAR = \frac{[Na^{+}]}{[(Ca^{2+} + Mg^{2+})/2]^{1/2}} \qquad (2.9)$$

Another term used to describe the exchangeable sodium in a soil is the exchangeable sodium percentage (ESP), where approximately:

$$ESP = ESR \times 100 \qquad (2.10)$$

Determining the cation exchange capacity (CEC) of a soil involves leaching the soil with a solution containing a replacement or index cation to be placed on all exchange sites. The soil is then washed, usually with alcohol, to keep it flocculated and to prevent loss of the index ion by hydrolysis, to eliminate soluble salts. The index cation is then extracted with another salt solution. The quantity of index cation in the final extract is a measure of the cation exchange capacity of the soil. The initial exchangeable cations of the sample can be determined by analyzing the cation concentration of the first leaching extract.

A second method of determining the cation exchange capacity of a soil involves repeated washings with an index cation. This consists of several cycles of adding the index cation, shaking, centrifugation, and decanting the supernatant solution. Analysis of the supernatant solution gives the amounts of individual exchangeable ions present.

The salt used to furnish the index cation should be relatively soluble in the alcohol used for sample washing. Sodium chloride is a common salt used to furnish the sodium index cation. An often overlooked source of error is that sodium chloride has a very low solubility in ethanol and it may precipitate during the sample washing and remain in the sample. The salt then dissolves in subsequent extraction steps leading to high CEC values. Okazaki et al. (1963) proposed a procedure which eliminates salt precipitation during washing. The anion of the salt solution is analyzed as well as the cation. The CEC then equals the quantity of index cations removed during extraction minus the quantity of anions. Determining the initial cation and anion concentrations of the salt solution eliminates further errors, and reliable CEC values are obtained.

2.6 Diffuse Double Layer

A second phenomenon, the diffuse double layer, coupled with the concept of cation exchange can be used to explain many properties of bentonite. Physical properties thus affected include Atterberg limits (Taylor, 1948, pp. 27-30) and hydraulic conductivity. Chemical properties include the type of cation adsorbed on the mineral surface. An understanding of diffuse double layer theory is therefore beneficial in any attempt to relate the engineering behavior of a specific clay to its physical and chemical properties.

The basic theory of the diffuse double layer (DDL) was developed by Gouy (1910, 1917) and by Chapman (1913) and is commonly referred to as the Gouy-Chapman theory (Bohn et al., 1979, p. 156). It rests upon four basic assumptions: (1) exchangeable cations exist as point charges; (2) colloid surfaces are planar and infinite in extent; (3) surface charge is distributed uniformly over the entire colloid surface; and (4) the static dielectric constant of the medium is independent of position. These assumptions are similar to those used by Debye and Hueckel (1923) to account for the effect of ion charge and concentration on the activity coefficient of an individual ion. Although these assumptions do not describe actual systems, the DDL theory works quite well for soil colloids.

The DDL theory relies upon the negative charge imparted to montmorillonite minerals due to broken chemical bonds, isomorphic substitution, or dissociated hydrogen from hydroxyl groups. The negatively charged clay particle creates an electric field. Exchangeable cations are attracted to and adsorbed onto the clay surface in order to satisfy the electric field. When the soil is dry, the exchangeable cations are assumed to be adsorbed directly on the colloid surface and are held very tightly. This electrostatic attraction follows Coulomb's law. The negative charge of the clay mineral and the layer of adsorbed exchangeable cations make up the Helmholtz double layer (Figure 2.4).

When water is introduced to the clay mineral, the adsorbed cations are no longer held as tightly and go into solution. The cation concentration is greatest near the mineral surface and has a tendency to diffuse into the lower cation concentration pore water. Cation diffusion tends

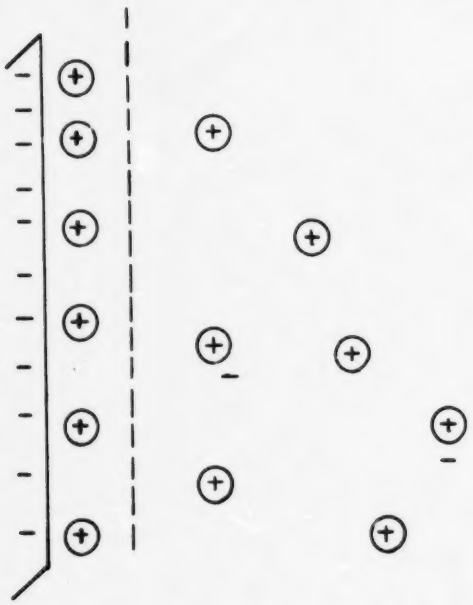

Figure 2.4 Schematic diagram of clay particle (left solid line) with adsorbed water layer (in between solid and broken line) and surface. Exchangeable cations in the diffuse ion-layer.

(Reprinted by permission of Elsevier Scientific Publishing Company, from R.N. Yong and B.P. Warkentin, Soil Properties and Behaviour, Figure 2.22, p. 58.
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to equalize concentration differences in the aqueous phase. Opposing cation diffusion is the negative electric field of the clay mineral resulting in the anion/cation distribution shown in Figure 2.5.

The cation concentration decreases away from the clay mineral surface while the anion concentration increases. The distance from the clay mineral surface where the solution concentration is affected by the clay mineral charge is called the diffuse double layer or double layer. Beyond the double layer, the cation and anion concentrations are equal. This region is defined as the bulk solution.

Increasing the bulk solution concentration reduces the tendency for cation diffusion away from the clay mineral surface, resulting in a decreased double layer thickness (Figure 2.6). The double layer thickness is also affected by cation charge. Increasing cation charge tends to reduce the double layer thickness, because fewer ions are required for charge neutralization. Higher charged cations are also held more tightly to the clay mineral surface.

The cation and anion distributions can be calculated from the Boltzman equation (Bohn et al., 1979, p. 157; Mitchell, 1976, p. 114):

$$\frac{C}{C_o} = \text{EXP}\ \frac{-Ze\psi}{kT} \qquad (2.11)$$

where C is the ion concentration at a specified distance from the clay mineral surface, C_o is the ion concentration in the bulk solution, Z is the ion valence, e is the electronic charge unit, ψ is the electrical potential of the clay mineral at the specified distance, k is the Boltzman constant, and T is the absolute temperature (Bohn et al., 1985, pp. 169-170)

Stern (1924) improved the model by dividing the region near the mineral surface in two. The layer of ions adsorbed on the mineral surface is known as the Stern layer. The second region consists of the diffuse or Gouy layer. The ions adsorbed in the Stern layer are subject to electrostatic and specific interaction. The Stern model assumes that some ions are tightly held at the mineral surface, thereby reducing the surface charge of the mineral. The diffuse portion then develops on a mineral surface having a reduced charge density. Figure 2.7 shows the Stern model distribution of three monovalent cations.

Figure 2.7 shows relatively few of the strongly hydrated lithium ions being adsorbed. Most of these are found in the diffuse layer. Potassium, a weakly hydrated ion, is adsorbed strongly to the mineral surface. Most of the potassium ions are found in the Stern layer. Moderately hydrated ions, such as sodium, have approximately equal concentrations in both the Stern and diffuse layers. Shainberg and Kemper (1966a,b) dealt extensively with the implications and applications of the Stern model and the consequences of its assumptions (Bohn et al., 1979, p. 161).

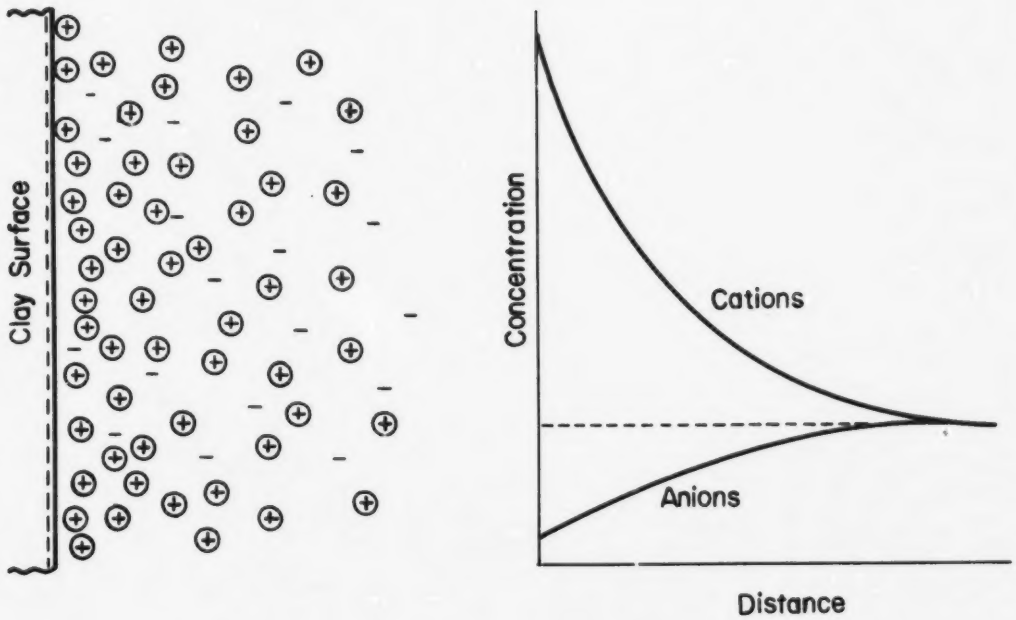

Figure 2.5 Ion distribution adjacent to a clay mineral surface according to the diffuse double layer theory.

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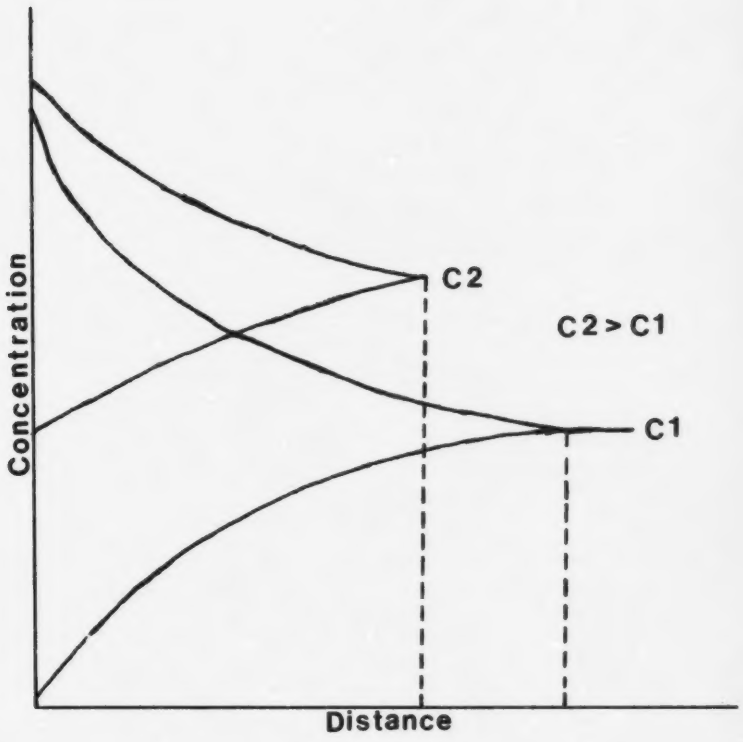

Figure 2.6 Effect of bulk solution concentration on double layer thickness. (from Nielsen et al., 1972)

(Adapted from Soil Water, 1972, pages 21-63, by permission of the American Society of Agronomy, Inc., and Soil Science Society of America, Inc.)

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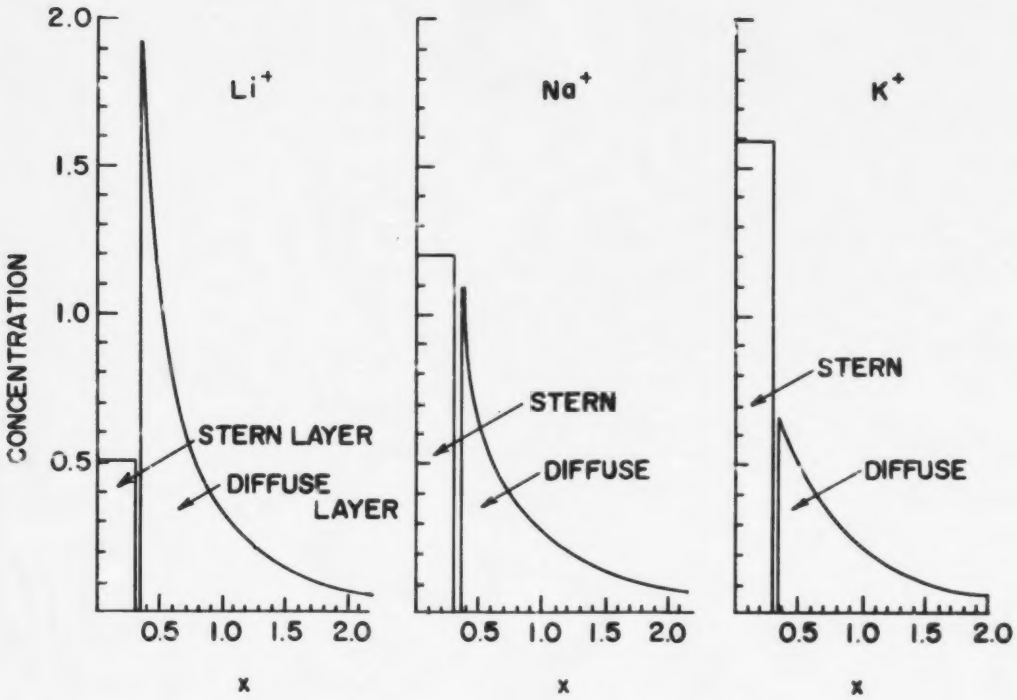

Figure 2.7 Cation distribution near a mineral surface for the Stern model (from Shainberg and Kemper, 1966b).

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The permeability of bentonite can be related directly to the double layer thickness (e.g. Yong and Warkentin, 1975, p. 151). The double layer thickness determines whether attractive forces contribute to particle aggregation while repulsive forces tend to create a dispersed, homogeneous mixture. Figure 2.8 shows the attractive and repulsive forces as a function of particle separation for three bulk solution concentrations.

The van der Waals attractive forces are insensitive to solution concentration, while the repulsive forces are very concentration-dependent. A high repulsive force prevents clay particles from coming into close proximity resulting in a stable, dispersed, homogeneous clay mixture. High repulsive forces dominate when the double layer is greatest. Attractive forces create clay aggregates resulting in an increase in the void ratio and thus relatively higher permeability values.

The extent of the double layer also influences parameters such as the plastic and liquid limits and swelling pressures of montmorillonite.

2.7 Mineral Swelling

Mineral swelling is also a function of interlayer bond strength and is inversely related to the interlayer bond energy. Swelling is also a function of the amount of mineral surface accessible to polar molecules such as water. Surface areas of layer silicates are related to their expansion properties and may be classified as external and internal. External surface refers to only the edges and faces of the whole crystal; internal surface refers to the area of basal plane surfaces. Nonexpanding minerals exhibit only external surfaces and have small surface areas. Expanding minerals exhibit both external and internal surfaces. Internal surface area is much larger than external area, resulting in very large total surface areas for expanding minerals such as montmorillonite. For example, the nonexpanding mineral kaolinite usually has a surface area of 10 to 20 x 10^3 m^2/kg, but the surface area of montmorillonite, an expanding mineral, can be as large as 800 x 10^3 m^2/kg (Bohn et al., 1985, p. 118; Mitchell, 1976, p. 39), of which up to 80% may be internal surface area.

Montmorillonite is classified as an expanding mineral because of its lower layer charge of 0.25 to 0.6 per formula unit. The c-spacing or the extent of swelling varies with the exchangeable cation and the degree of interlayer solvation. Complete drying yields a spacing of 0.95 to 1.0 nm. Full hydration can swell the layer to tens of nanometers. Swelling is likely to be a significant factor affecting the hydraulic conductivity of bentonite (e.g. Mitchell, 1976, pp. 349-351).

2.8 Properties of Bentonite Used in This Study

The bentonite used is American Colloid Company Volclay tablets. American Colloid Company describes the tablets as pre-formed compressed tablets made of high swelling sodium bentonite having a 2.5 cm (1/2

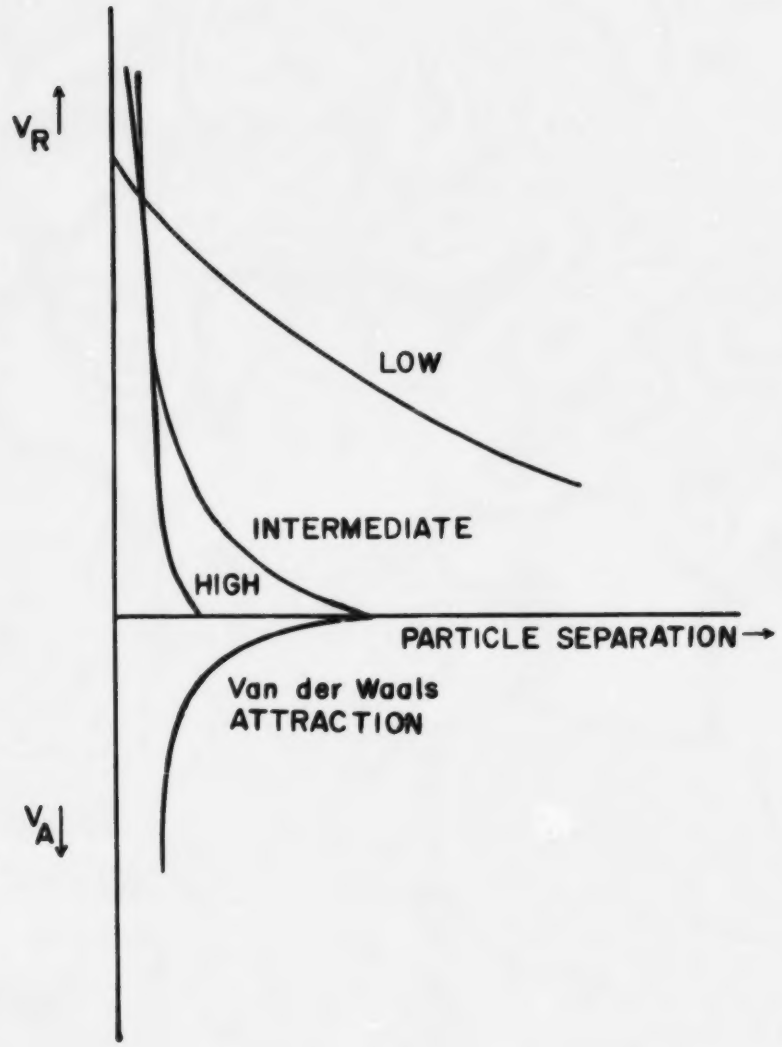

Figure 2.8 Attractive (V_A) and repulsive (V_R) energy as a function of particle separation for three electrolyte concentrations.

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inch) diameter. The bentonite tablets are comprised of a hydrous silicate of alumina having a minimum montmorillonite content of 90%. Other minerals include biotite, feldspar, and selenite. The pH ranges from 8.5 to 10.5. Individual tablet density averages 129 lb/ft^3 (2.07 g/cm^3). Dry bulk density averages 82 lb/ft^3 (1.31 g/cm^3). Specific gravity ranges from 2.3 to 2.5, which implies that actual grain density ranges from 143.5 lb/ft^3 to 156 lb/ft^3.

American Colloid tests show that for 72 g of 1.25 cm (1/2-inch) diameter tablets the total volume is 83.4 cm^3, pore volume is 49.9 cm^3, and the volume of solids is 33.5 cm^3. Using the relationship

$$\rho_s = M_s/V_s \tag{2.12}$$

where ρ_s is the mean particle density, M_s is the mass of solids, and V_s is the volume of solids, the mean particle density of the tablets is 134.2 lb/ft^3 (2.15 g/cm^3). Comparing the individual tablet density supplied by the manufacturer with the grain density calculated from the specific gravity of the tablets suggests a tablet porosity ranging from 10 to 17 percent by volume. Using the relationship

$$e = n/(1 - n) \tag{2.13}$$

where e is the void ratio and n is the porosity, a void ratio ranging from 0.11 to 0.20 is calculated for the individual tablets.

The moisture content of the dry tablets was determined in the laboratory. Three sets of ten tablets were weighed, oven-dryed, and reweighed. An average moisture content of 11% was determined. The manufacturer of the tablets estimated a tablet moisture content of 10%.

A whole rock analysis of the tablets was performed by Copper State Analytical Lab, Inc., of Tucson, Arizona. Two samples, representing two delivery dates, were submitted for analysis. Sample One represents the most recently received batch; Sample Two was received approximately eleven months earlier. Results of the whole rock analysis are summarized in Table 2.1. With the exception of the SiO_2 determination, all analyses utilized atomic absorption spectroscopy. SiO_2 values were determined by gravimetric methods. All values are reported as percentages and expressed as oxides.

The sodium oxide values are much greater than those of calcium oxide, which is indicative of a sodium bentonite. The reported SiO_2 values range from 70 to 76%, the Al_2O_3 values from 8 to 16%. Grim and Guven (1978, p. 18) report SiO_2 values ranging from 41 to 81% and Al_2O_3 values from 0.14 to 27%. The chemical composition of the bentonite used here falls within the wide range of values reported by Ross and Hendricks (1945) for bentonites from different sources.

Table 2.1 Summary of Whole Rock Analysis for Bentonite Samples

Constituent	Sample #1	Sample #2
SiO_2	69.8	76.2
Al_2O_3	15.9	8.1
CaO	0.21	0.40
MgO	1.96	3.42
Fe_2O_3	5.29	4.58
Na_2O	3.3	3.6
K_2O	1.06	0.84
P_2O_5	0.068	0.080
TiO_2	0.19	0.20
MnO	0.12	0.14
LiO	1.86	2.10

CHAPTER THREE

PERMEANT CHARACTERISTICS THAT AFFECT THE HYDRAULIC CONDUCTIVITY OF A POROUS MEDIUM

3.1 Introduction

Permeant chemistry can affect the hydraulic conductivity of a porous medium, especially when it contains clay, by influencing clay dispersion, flocculation, and swelling, e.g. by affecting the type and amount of cations in the mineral interlayers.

Permeant influence can be divided into its chemical and its physical characteristics. The chemical characteristics include pH, total salt concentration, specific cation concentrations, and organic matter content. The total salt concentration and the specific cation concentrations will be discussed later. The influence of permeant organic matter on the hydraulic conductivity of porous media is beyond the scope of this report. Physical characteristics of the permeant include color and turbidity. Color can be an indicator of the organic matter content of a water. Turbidity, the amount of suspended matter in a water, is the only physical permeant characteristic examined in this report.

The major review paper by Neuzil (1986) on groundwater flow in low-permeability environments places the two factors addressed here within a broader context of nonhydraulic parameters influencing flow. The paper includes, in particular, an extensive list of references on ultra-filtration, or ion removal, during solution flow through bentonite.

3.2 Chemical Characteristics

Many researchers have studied the effects of permeant chemistry on soil hydraulic conductivity. Among the earliest were Fireman and Bodman (1939). They studied the effects of distilled water and of a saline water on the hydraulic conductivity of two soils. Only the results obtained from the montmorillonite soil are reported here. The saline water had a total concentration of 69 milliequivalents per liter (meq/ℓ) and a sodium adsorption ratio (SAR) of 27. The hydraulic conductivity obtained with the saline water was much greater than that obtained with distilled or salt-free water. Fireman and Magistad (1945) found that hydraulic conductivities of five western United States soils decreased when water with a 25 meq/ℓ concentration and SAR's ranging from 4.5 to 31.5 was replaced by a water with a concentration of 4.4 meq/ℓ and a SAR of 1.5. They concluded that water low in salts will cause substantial decreases in hydraulic conductivity if earlier percolating water had high total salt concentrations. This is especially so if the previous salts contained sodium. These findings agree with the permeability implications of the diffuse double layer theory (Section 2.6), and the development of immobilized water layers (e.g. Yong and Warkentin, 1975, p. 151), although this is a

controversial explanation, at best (Mitchell, 1976, p. 351; Neuzil, 1986).

Henderson (1958) tested four soils using permeants of varying salt concentrations and having SAR's less than 20. Hydraulic conductivity decreased as the total salt concentration decreased. Extreme hydraulic conductivity reductions occurred when low sodium waters (SAR $\leq$ 10) having a total concentration from 0 to 10 meq/ℓ were used. Large hydraulic conductivity decreases were also observed with waters having higher SAR values and salt concentrations greater than 10 meq/ℓ.

Others have found similar reductions in hydraulic conductivity over a wide range of permeant concentrations and SAR levels. McNeal (1965) and McNeal and Coleman (1966) determined the hydraulic conductivity of seven soils using permeants having SAR's ranging from 0 to ∞ and total salt concentrations of 3.1, 12.5, 50, 200 and 800 meq/ℓ. The soils had montmorillonite contents ranging from 1 to 40%. They found large reductions in hydraulic conductivity in five of the soils when tested with permeants having SAR values under 25 and total salt concentrations under 50 meq/ℓ.

Naghshineh-Pour et al. (1970) determined the hydraulic conductivity of four soils using permeants having SAR values of 0, 10, 19, 27 and 38 and total salt concentrations of 10.5, 54, 107, and 300 meq/ℓ. Two of the soils had montmorillonite contents greater than 40%. The other two consisted primarily of halloysite and kaolinite. Only the results for the montmorillonitic soils are discussed here. A liter of the 300 meq/ℓ solution of a given SAR value was initially percolated through a soil column. This was followed by a liter and a half of the more dilute solutions, from 107 to 10.5 meq/ℓ, of the same SAR. The flow rate was measured using the last half liter of solution. A rapid decrease in hydraulic conductivity was observed for increasing SAR values and decreasing solution concentration. The fastest change occurred at a salt concentration below 54 meq/ℓ and above a SAR value of 10. Pronounced swelling and dispersion were observed at high SAR and low salt concentrations, indicated by increases in soil volume of 28 to 33 percent.

Reeve and Tamaddoni (1965) studied the effects of high solution concentrations at varying SAR values on the hydraulic conductivity of a clay loam containing a 15% expanding lattice-type clay. Permeants having SAR values of 0, 80, 180 and ∞ and total salt concentrations ranging from 63 to 4000 meq/ℓ were used to determine soil hydraulic conductivity values. Solutions of varying concentration but with constant SAR values were applied successively to a given soil sample. Two application methods were employed. The first, known as the high series, subjected the sample to the high concentration solution followed by the lower concentration solutions. The second method, known as the low series, subjected the sample first to the low concentration solution, followed by the higher concentration solution. The hydraulic conductivity was a function not only of the solution composition, but also of the initial solution concentration. If the initial solution concentration was low, conductivity at all later concentrations was lower than if high initial concentrations were used.

Varallyay (1974) investigated three sets of soil samples. Two included a significant montmorillonite fraction. Considerable and rapid descreases in hydraulic conductivity were observed during flow testing with distilled water. ESP was found to be a dominant factor in hydraulic conductivity, at least up to a certain threshold value for either clay or sodium content.

Quirk (1952; 1968) investigated the differences in behavior of sodium and calcium saturated montmorillonite clays. He found a uniform flow rate through sodium saturated montmorillonite using a 0.5 M NaCl solution. When the solution was replaced with a 0.25 M NaCl solution, the clay swelled and became virtually impermeable. Norrish and Quirk (1954) attributed this behavior to crystalline swelling of montmorillonite. A montmorillonite flake in a 0.5 M NaCl solution gave an X-ray c-spacing of 19 Å. At a concentration of 0.35 M the spacing increased to 40 Å. For calcium montmorillonite the spacing never increased beyond 19 Å.

Quirk and Schofield (1955) studied the flocculation of clays. They found results similar to those of Mattson (1929), who reported that a 0.1 M NaCl solution was required to flocculate a stable montmorillonitic suspension, even though only a 0.02 M NaCl solution was required to flocculate montmorillonite. The differences in required solution concentration were attributed to the presence of organic matter.

McNeal et al. (1968) evaluated the role of soil texture on hydraulic conductivity in the presence of mixed-salt solutions. Soils having variable clay content, but nearly uniform clay-fraction mineralogy, were studied. The hydraulic conductivity decreased with increasing clay content and SAR values. In general, the hydraulic conductivity was strongly dependent on soil texture, with high-sodium, low-salt solutions having a greater effect on the conductivity of soils having greater clay contents.

Early investigators studying the effects of solution chemistry on the hydraulic conductivity of soils concluded that waters having high SAR values and low salt concentrations very effectively reduce soil hydraulic conductivities. It was also recognized that changes in soil hydraulic conductivity were not only a function of the solution chemistry, but of several variables, including clay content and type of clay minerals present. Swelling and dispersion were recognized as major processes in reducing soil hydraulic conductivities. Shainberg and Caiserman (1971) assessed the relative importance of swelling and dispersion in decreasing the hydraulic conductivity of clays and soils. In the first of two sets of experiments, the movement of clay particles was greatly restricted so that swelling would be the dominant process affecting the hydraulic conductivity. In the second, water movement was affected by dispersion and particle movement in addition to swelling. The relative importance of swelling and dispersion was assessed by comparing the effect of solution composition on the rate of water movement through the clay samples. Intrinsic permeabilities calculated using the Kozeny-Carman equation were higher than experimental values for sodium montmorillonite and lower for calcium

montmorillonite. Shainberg and Caiserman (1971) quote Lagerwerff et al. (1969), who suggested that a correction factor for adsorbed water, with less mobility than non-adsorbed water, be incorporated into the Kozeny-Carman equation. Kemper et al. (1964) measured the mobility of water adjacent to clay surfaces and estimated the first molecular layer of adsorbed water on sodium and calcium clays to have mobilities of 30% and 5%, respectively, of the bulk water. Agreement between predicted and experimental values of the Shainberg and Caiserman (1971) results was very good when the correction factor was used. The authors concluded that the mechanism for hydraulic conductivity decreases in sodium montmorillonite is the closing of conducting pores by swelling. They also showed that the hydraulic conductivity of the clay, as a function of ESP, depends on the direction from which equilibrium is approached. The measured hydraulic conductivity values obtained when approaching equilibrium from the calcium end (SAR $\rightarrow$ 0) were higher than those obtained when equilibrium was approached from the sodium end (SAR $\rightarrow \infty$).

Two mechanisms have been suggested as possible explanations for this direction dependency called hysteresis. One assumes hysteresis in maintaining ionic equilibrium. If equilibrium is obtained from the sodium end, a higher percent of adsorbed sodium is present in the c-spacing, resulting in lower hydraulic conductivity. If equilibrium is attained from the calcium end, the adsorbed sodium in the c-spacing is reduced, due to the affinity of montmorillonite for calcium, resulting in higher hydraulic conductivity values (Shainberg and Kemper, 1966).

A second mechanism is that the ESP of the clay in equilibrium with any solution is independent of experimental conditions and the hysteresis loop is due to the dependency of the hydraulic conductivity on the initial conditions. Calcium montmorillonites are stable and have large pores. They exist in packets, or tactoids, averaging four to five clay platelets each (Blackmore and Miller, 1961). Van Olphen (1977, p. 48) defines tactoids as spindle-shaped regions in which the particle concentration is higher than in the bulk of the colloidal suspension. Frenkel et al. (1978) state that calcium-saturated montmorillonite clay particles commonly consist of packets or tactoids of four to nine clay platelets arranged parallel to each other at distances of 9Å. Sodium introduced to calcium montmorillonites is concentrated on the external surfaces of the tactoids, affecting the size and distribution of the pores only slightly. Eighty to ninety percent calcium solution will form tactoids, but with fewer platelets in a tactoid resulting in lower hydraulic conductivity values.

Shainberg and Caiserman (1971) showed that a slight addition of exchangeable sodium to a calcium saturated clay paste has a moderate effect on the water movement within the paste. The hydraulic conductivity of the calcium saturated clay paste decreased significantly when the ESP was increased from 0 to 25. Above 25, the hydraulic conductivity of the paste is identical to that of a pure sodium montmorillonite. The ESP also affects the mobility of montmorillonite particles (BarOn et al., 1970). Particle size and swelling are not affected by a low ESP; therefore, introducing a low

percentage of sodium into the c-spacing of a calcium montmorillonite strongly increases the mobility of the clay particles. Water movement through the clay moves the particles which block the large conducting pores.

Frenkel et al. (1978) determined the hydraulic conductivities and gradients along soil columns packed with three soil types adjusted to different ESP values. The montmorillonitic soils varied in clay content from 2.9 to 18.0%, and had a uniform clay mineralogy consisting of 42% montmorillonite, 29% mica, 16% quartz and feldspar, and 13% other species. Significant reductions in hydraulic conductivity were observed with increasing ESP at a given solution concentration. When the solution was changed to distilled water, hydraulic conductivities decreased markedly at clay contents greater than ten percent, and at all ESP values. For clay contents of less than ten percent, reductions only occurred for ESP values of 20 and 30. When distilled water was percolated through the soil, clay particles appeared in the effluent.

Reductions in hydraulic conductivities were also observed for montmorillonitic soils which are not expected to exhibit appreciable swelling. Swelling is not generally appreciable unless the ESP exceeds 25 or 30 (see also Shainberg and Caiserman, 1971). Dispersion, however, can occur at ESP levels as low as 10 to 20, if the solution concentration is less than 10 meq/ℓ (Felhendler et al., 1974, as quoted by Frenkel et al., 1978). Reductions in hydraulic conductivities under these conditions were attributed to particle dispersion and pore plugging. When leached with a high concentration solution, the hydraulic conductivity did not increase as would be expected if swelling and shrinkage of pores were the cause of the reduced hydraulic conductivity. Clay dispersion and plugging also caused reduced hydraulic conductivity in soils having a higher clay content and ESP.

Frenkel et al. (1978) concluded that dispersion appears to be the main cause of reduced hydraulic conductivities of montmorillonitic soils at high ESP (30%) and high clay content (18%). Dispersion and plugging are intensified with increased ESP and clay content and reduced solution concentration. Coarse textured soils do not generally experience plugging because of the relatively large pores and fast water velocities.

Rowell et al. (1969) evaluated the hydraulic conductivity changes and the swelling process for a soil containing montmorillonite for a range of ESP and solution concentrations. They were primarily interested in answering three questions: 1) Are there two solution concentration ranges, the higher range causing swelling only and the lower causing both swelling and dispersion; (2) Does a direct relationship exist between swelling and hydraulic conductivity at the high concentration; and (3) How important is mechanical stress in determining the concentration at which dispersion occurs? The soils they tested contained approximately 22 percent clay. The clays were predominately illite (75-85%) with some kaolinite (10-15%) and montmorillonite (5-10%). Six solutions having different concentrations were percolated through the soils. The most concentrated solution was percolated through first, followed by the more dilute solutions. The results

indicate that hydraulic conductivity begins to decrease at the same solution concentration as that at which the clay begins to swell. Changes in the permeability are directly controlled by the swelling of the clay until clay dispersion and movement begins. The results also suggest that dispersion depends on mechanical stress. When small mechanical stresses are applied, the proportion of the clay which swells and disperses depends directly on the ESP. Large mechanical stresses may disperse most of the clay, even at low ESP values.

The dependence of dispersion on the applied mechanical stress is based on the theory of colloid stability presented by Verwey and Overbeek (1948, Chapters VII, XII), which suggests that when a swollen clay is in equilibrium with an electrolyte solution, the clay particles are in a position of minimum potential energy. A potential energy barrier, which decreases with reductions in electrolyte concentration, prevents the clay particles from moving away from each other. A mechanical stress applied to the clay can push the clay particles over this barrier, resulting in dispersion. The larger the stress, the larger the barrier that can be overcome, and, therefore, the higher the electrolyte concentration at which dispersion can occur. A pressure must be applied to the dispersed particles to reflocculate them.

Dispersion was also found to depend on the soil ESP value. As the sodium percentage is reduced, less swelling and dispersion occurs. Glaeser and Mering (1954) and Mungan and Jessen (1963) have suggested that clays separate or "de-mix" into calcium and sodium saturated zones. The relative amounts of each depend upon the soil ESP value. The sodium saturated zones would swell, resulting in dispersion when the turbidity concentration was reached. The unswollen calcium saturated zone would not disperse.

Rowell et al. (1969) suggest that, in contrast to the observations on turbidity of percolates, large mechanical stresses may cause undispersed clay to disperse regardless of the exchangeable sodium present in the clay. They suggest that extending the hypothesis of Glaeser and Mering would indicate that the unswollen calcium saturated zones can also be dispersed, given a large enough stress. A large mechanical stress would also overcome the potential energy barrier.

3.3 Physical Characteristics

Particulate matter in permeants can have a significant effect on the hydraulic conductivity of a porous medium, especially a fine-grained one. Particles in water are usually defined as finely divided solids, larger than molecules, but generally not discernible by the unaided eye. Natural sources of particulate matter include soil weathering and biological activity. Clays are the principal by-products of weathering. Algae, bacteria, and microorganisms are the principal particles produced biologically. Particles in water may clog pores, thereby reducing the hydraulic conductivity. Particles may also serve as adsorption sinks, resulting in a larger particle.

Hunt et al. (1987) examine theories and experimental data relating to colloid migration in an effort to quantify colloid migration in porous

media. They recognize that most of the quantitative work has been undertaken in the field of water filtration, where deep bed granular media filters are used for particulate removal. The following paragraphs, extracted from Hunt et al. (1987) identify some salient aspects of particle effects on water flow.

Migration of particles in porous media and subsequent reduction of hydraulic conductivity depends on the ability of the particle to infiltrate the media surface. If the particle diameter is larger than the media pore diameter, geometric straining occurs. Sakthivadivel (1969) and Sherard et al. (1984) established that straining will occur if the ratio of the particle diameter, d_P, to the media diameter, d_M, is greater than 1/9. Herzig et al. (1970) have suggested that very little straining will occur if $d_P < d_M/50$. Particles which penetrate the media surface would be retained within the pores only under favorable media-particle attachment conditions or if the particles were retained within quiescent pore spaces (Krone et al., 1958; Hunter and Alexander, 1963).

Several mechanisms affect particle collisions with porous media. Brownian motion dominates for particles smaller than a few micrometers. Larger particles flowing with streamlines can collide with media surfaces through interception. Particles with a density different from that of the fluid can cross streamlines and collide due to gravitational sedimentation.

Particle collisions with a medium do not ensure attachment. Particle-media interactions must also be favorable. Hydrodynamic, London-van der Waals, and electrostatic forces influence particle-media attachment. Hydrodynamic force results from squeezing out fluid as particles approach the media surface. London-van der Waals force is a short range attractive force. Electrostatic repulsion occurs if the particles and media have the same charge. The magnitude of the repulsion may be overcome if the surrounding water has a high ionic strength. High ionic strengths can compress the diffuse double layer and reduce the distance over which electrostatic repulsion forces act. London-van der Waals forces may then dominate and attachment is favored.

If particle-media attachment is favorable, particles occupy a part of the pore space, resulting in decreased hydraulic conductivity. The deposited particles act as collectors for other particles, resulting in further pore space reductions. Under steady flow conditions, fluid velocity increases with a reduction in pore volume. The higher velocity hinders particle deposition, resulting in an eventual equilibrium.

Particulate matter in water can be quantified by turbidity readings or by particle counts. Turbidity is an optical property of the sample causing light to be scattered and adsorbed, rather than transmitted (APHA, 1980, p. 131). One method to measure the turbidity of a water uses nephelometry to measure the intensity of light scattered by turbidity particles. The sample is illuminated and photoelectric

detectors with a readout device indicate the intensity of scattered light at right angles to the light path. A formazin polymer suspension is generally used as a standard. Turbidity measurements obtained in this manner are expressed in nephelometric turbidity units (NTU).

A second method to quantify particulate matter is by particle concentration measurements. These provide information on particle size distribution in a sample. Particle counter sensors are available in different size ranges. Common ranges include 1.0 to 60 μm and 2.5 to 150 μm. Particle counts may be adjusted to measure particles in about 12 subranges within the chosen micrometer sensor range.

3.4 Characteristics of the Permeants Used in This Study

Five permeants were used to determine how solution chemistry affects the hydraulic conductivity of bentonite. These included a groundwater from the Ogallala aquifer in northern Texas, distilled water, a high-concentration calcium water, a low-concentration calcium water, and a low-concentration sodium water.

The chemical composition of the Ogallala groundwater is given in Table 3.1. The analysis was conducted at the Environmental Engineering Laboratory at the University of Arizona. The table includes an earlier analysis of Ogalla groundwater conducted by Cagel and Baldwin (1978). The values compare favorably. The groundwater turbidity was determined using the particle count method. The particle size distribution is shown in Table 3.2. Three 10 ml samples were analyzed. Nancy Patania, a graduate student in the Environmental Engineering Program at the University of Arizona, suggested that the number of particles reported in Table 3.2 may not reflect the total number of particles present when the groundwater was originally obtained. Experiments conducted by graduate students in the Environmental Engineering Program indicate that significant adsorption of particles onto plastic occurs in a relatively short time. An original particle count of 100,000 was reduced to 40,000 after storing the water for two weeks in a plastic container. The Ogallala groundwater was stored for approximately three months in five gallon plastic containers before the particle distribution count was performed.

The distilled water used in this study was obtained from the Chemical Engineering Department at the University of Arizona. Chemical analysis showed it to be free of salts. The water had a much smaller particle count than the Ogallala groundwater (Table 3.2). Three 10 ml samples were analyzed. The reservations expressed by Olson and Daniel (1981) in their comprehensive review of hydraulic conductivity measurements about the use of distilled water are recognized, as well as their preference for the use of tap water. While this preference may be fully justified for measurements intended to directly simulate particular field conditions, it is believed that testing with distilled water should provide a more consistent reference basis.

The 1000 ppm calcium water was made by dissolving 18.34 g of calcium chloride dihydrate in 5 liters of distilled water. A calcium determination showed 980 ppm calcium concentration. The fourth

permeant was a 45 ppm calcium water obtained by dissolving 1.65 g of calcium chloride dihydrate in 10 liters of distilled water. The fifth permeant was a 39 ppm sodium water obtained by dissolving 0.991 g of sodium chloride in 10 liters of distilled water.

Table 3.1 Average Chemical Composition of Ogallala Ground Water

Component	Concentration (ppm)	
	This Study	Cagel and Baldwin (1978)
Na^{+}	37	39
Ca^{2+}	43	45
Mg^{2+}	33	29
K^{+}	--	2
Cl^{-}	--	23
SO_4^{2-}	--	41
HCO_3^{-}	280	294
pH	7.5	7.9
TDS	360	379
DO	--	8
SiO_2	--	46

Table 3.2 Average Particle Distribution Count of Permeant Waters

Diameter Range (μm)	Average Number of Particles per 10 ml Sample
	Ogallala Groundwater:
1.0 - 2.5	3972
2.5 - 5.0	3001
5.0 - 10.0	1322
10.0 - 20.0	246
20.0 - 40.0	21
40.0 - 60.0	1
Total:	8565
	Distilled Water:
1.0 - 2.5	593
2.5 - 5.0	188
5.0 - 10.0	34
10.0 - 20.0	10
20.0 - 40.0	1
40.0 - 60.0	0
Total:	826

CHAPTER FOUR

HYDRAULIC CONDUCTIVITY TESTING THEORY

4.1 Introduction

Conventional laboratory methods to determine the hydraulic conductivity of soils include the falling head test, the constant head or constant pressure injection test, and tests which utilize soil consolidation or oedometer data. The falling head test is usually considered limited to relatively pervious, saturated soils, while the oedometer tests are usually reserved for low permeability clayey soils. The constant head test is widely used on all types of soils (Lambe and Whitman, 1969, p. 281), although ASTM restricts its use to soils containing not more than 10% soil passing the 75-μm (No. 200) sieve in order to limit consolidation influences (ASTM D 2434-68).

Transient pulse testing (e.g. Brace et al., 1968; Lin, 1977; Hsieh et al., 1981; Neuzil et al., 1981) has two potentially important advantages over conventional laboratory tests utilized to determine the hydraulic conductivity of a sample of low permeability. Constant head testing may require extensive time periods before representative hydraulic conductivity values are obtained. Transient pulse testing may provide representative hydraulic conductivity values in relatively short time periods. A second potential advantage of transient pulse testing is its capability of providing a storage value. Disadvantages of this method include considerable remaining uncertainty about its applicability for highly deformable materials such as clays, and even more so for swelling bentonites. The comprehensive review by Neuzil (1986) of flow through low-permeability media includes a discussion of the comparative merits of steady-state and transient testing, as well as of consolidation testing of highly deformable media such as soft clays. The likely necessity for corrections to account for large deformations is pointed out.

4.2 Constant Head Test Theory

The constant head test is based on laboratory experiments conducted by Darcy (1856). The rate of fluid flow, Q (L^3/t), is proportional to the cross-sectional area of the sample, A (L^2), the change in hydraulic head across the sample, $h_2 - h_1$ (L), and inversely proportional to the sample length , L (L):

$$Q = -KA[(h_2 - h_1)/L] \qquad (4.1)$$

K is the hydraulic conductivity. The hydraulic gradient $[(h_2 - h_1)/L]$ describes the change of piezometric head per unit length of sample.

Darcy's law is valid for linear-laminar flow in a porous medium. At

high flow velocities, the inertial forces in the fluid begin to dominate, resulting in turbulent flow. In turbulent flow, head loss increases exponentially with velocity. Reynolds number, R_e, a dimensionless number expressing the ratio of inertial to viscous forces acting on a fluid, is used to distinguish between laminar flow and turbulent flow. The Reynolds number has been developed for pipe flow applications, but a Reynolds number for flow through porous media has been defined as (Bear, 1979, p. 65; Freeze and Cherry, 1979, pp. 72-73):

$$R_e = (qd)/\nu \qquad (4.2)$$

where q = specific discharge = Q/A, $[L/t]$
d = a representative length of the porous media, $[L]$
ν = kinematic viscosity of the fluid, $[L^2/t]$

It is customary to use the mean grain diameter, d_{50}, as the representative length of the porous media. Bear (1979, pp. 65-66) and Freeze and Cherry (1979, pp. 72-73) suggest that the linear-laminar flow regime has an upper limit defined by Reynolds numbers not exceeding 1 to 10.

Swartzendruber (1962) and Bolt and Groenevelt (1969) suggest that Darcy's law may not be valid for very low flow rates in fine-grained low permeability materials. A threshold hydraulic gradient may exist below which flow does not take place. There is no agreement on the mechanism, and the experimental evidence is still open to some doubt (Freeze and Cherry, 1979, p. 72). The application of Darcy's law to fluid flow in fine-grained materials such as bentonite could be problematic. If a lower limit to Darcy's law does exist, hydraulic conductivities of fine-grained, low permeability materials determined from traditional testing methods may be questionable. Nevertheless, standard laboratory testing practice continues to use applications of Darcy's law in determining the hydraulic conductivity of clays (Lambe and Whitman, 1969, p. 281; Freeze and Cherry, 1979, pp. 337-338; and Holtz and Kovacs, 1981, pp. 380-385). Mitchell (1976, pp. 349-351), after reviewing the available evidence, concludes that Darcy's law is valid. Neuzil (1986) in a more recent and far more comprehensive review concludes that "... the case for observed non-Darcian behavior in experimental data is weak ... the use of Darcy's law to describe flow under these conditions represents a hypothesis; to be on firm analytical ground, it should be tested." There is less agreement on which test is most appropriate for low permeability clays. Klute (1965) recommends the constant head test for samples having conductivities greater than 0.01 cm/min and the falling head test for samples with lower conductivity. Lambe and Whitman (1969, p. 281) recommend that the falling head test be limited to pervious soils, and they state that the constant head test is widely used on all types of soils.

The hydraulic conductivity is a function of both fluid and medium properties. Influencing fluid properties include density and viscosity. Important medium properties include grain size distribution, grain shape, tortuosity, specific surface, and porosity.

4.3 Transient Pulse Testing Theory

4.3.1 Introduction

The primary objective of transient pulse testing of bentonite is to evaluate the applicability of an alternative method for determining the hydraulic conductivity of the clay. This measurement is difficult since standard laboratory techniques require steady flow and saturated conditions. For low permeability materials, long periods of time are required to assure saturation and to establish steady flow. Bentonite permeability testing is further complicated by the simultaneous occurrence of consolidation and swelling.

Hsieh et al. (1980) review transient pulse testing and analysis methods. They present a general solution and develop limiting cases. Neuzil et al. (1981) present a graphical method for analyzing data from a transient pulse test to obtain the hydraulic properties of the sample.

4.3.2 Mathematical Analysis of the Transient Pulse Test

Brace et al. (1968) developed a transient flow model to measure the hydraulic conductivity of a granite. Their apparatus consisted of a rock sample sandwiched between two fluid reservoirs. At the start of the experiment, the fluid pressure in the upstream reservoir is increased. Fluid flows from the upstream reservoir, through the sample, and into the downstream reservoir, resulting in a pressure decay in the upstream reservoir and a pressure build-up in the downstream reservoir. The hydraulic conductivity of the sample is calculated from the pressure decay in the upstream reservoir. The method assumes no compressive storage in the sample. This may be valid for crystalline rock. It breaks down for materials with significant porosity and compressive storage, such as bentonite.

Lin (1977) developed a transient pulse test which accounted for compressive storage in the sample. The model assumes that the compressive storage of the sample is known. In many cases the storage term is unknown, making it necessary to independently measure the bulk compressibility and porosity of the sample.

Hsieh et al. (1981) developed a model which accounts for the compressive storage of a sample without requiring that it be known, using a mathematical model designed by Brace (1968) and presented by Lin (1977). The equation describing one-dimensional flow of a slightly compressible fluid in a saturated porous medium with constant hydraulic conductivity K and specific storage S_s is given by (Hsieh et al., 1981):

$$\frac{\partial^2 h}{\partial x^2} - \frac{S_s}{K}\frac{\partial h}{\partial t} = 0 \qquad \text{for } 0 < x < \ell \text{ and } t > 0 \qquad (4.3)$$

where h is the hydraulic head in the sample, x is the distance along the sample, with x = 0 at the downstream face, and x = ℓ at the

upstream face. t is the time from the start of the experiment. At the start of the experiment, hydraulic head in the sample is uniformly 0:

$$h(x,0) = 0 \quad \text{for} \quad 0 \leq x \leq \ell \tag{4.4}$$

The upstream and downstream faces of the sample are in direct contact with the respective reservoirs, giving:

$$h(o,t) = h_d(t) \quad \text{for} \quad t \geq 0 \tag{4.5}$$

and

$$h(\ell,t) = h_u(t) \quad \text{for} \quad t \geq 0 \tag{4.6}$$

where h_d is the hydraulic head in the downstream reservoir and h_u is the hydraulic head upstream.

Conservation of mass at the sample reservoir interface is expressed by (Hsieh et al., 1981):

$$\frac{S_d}{KA}\frac{dh_d}{dt} - \left(\frac{\partial h}{\partial x}\right)_{x=0} = 0 \quad \text{for} \quad t > 0 \tag{4.7}$$

and

$$\frac{S_u}{KA}\frac{dh_u}{dt} + \left(\frac{\partial h}{\partial x}\right)_{x=\ell} = 0 \quad \text{for} \quad t > 0 \tag{4.8}$$

where S_d is the compressive storage of the downstream reservoir, and S_u the compressive storage of the upstream reservoir. At the start of the test, the hydraulic head in the downstream reservoir equals the head in the sample:

$$h_d(0) = 0 \tag{4.9}$$

An instantaneous head H is applied at the start of the test to the upstream reservoir:

$$h_u(0) = H \tag{4.10}$$

The solution of Equations (4.3) through (4.10) contains a dimensionless time α:

$$\alpha = \frac{Kt}{\ell^2 S_s} \tag{4.11}$$

and the ratio β of compressive storage in the sample to the compressive storage in the upstream reservoir:

$$\beta = \frac{S_s A\ell}{S_u} \tag{4.12}$$

as well as γ, the ratio of the compressive storage in the downstream reservoir to the compressive storage in the upstream reservoir:

$$\gamma = \frac{S_d}{S_u} \tag{4.13}$$

The compressive storage of the fluid reservoirs is the sum of the compressibility of the fluid in the reservoir and of the reservoir itself, including all tubing. The compressive storage of the reservoirs must be determined experimentally.

The general solution for dimensionless hydraulic head in the upstream reservoir is (Hsieh et al., 1981)

$$\frac{h_u}{H} = \frac{1}{1+\beta+\gamma} + 2\sum_{m=1}^{\infty} \frac{\exp(-\alpha\phi_m^2)(\beta+\gamma^2\phi_m^2/\beta)}{\left[\frac{\gamma^2\phi_m^4}{\beta^2} + (\gamma\beta+\gamma^2+\gamma+\beta)\frac{\phi_m^2}{\beta} + (\beta^2+\gamma\beta+\beta)\right]} \tag{4.14}$$

where ϕ_m are roots of

$$\tan\phi = \frac{(1+\gamma)\phi}{\frac{\gamma\phi^2}{\beta} - \beta} \tag{4.15}$$

The dimensionless hydraulic head in the downstream reservoir is given by (Hsieh et al., 1980):

$$h_d = \frac{1}{1+\beta+\gamma} + 2\sum_{m=1}^{\infty} \frac{\exp(-\alpha\phi_m^2)(\beta-\gamma^2\phi_m^2/\beta)}{\left[\frac{\gamma^2\phi_m^4}{\beta^2} + (\gamma\beta+\gamma^2+\gamma+\beta)\frac{\phi_m^2}{\beta} + (\beta^2+\gamma\beta+\beta)\right]\cos\phi_m} \tag{4.16}$$

where ϕ_m are the roots of equation 4.15.

An approximate solution for small α, known as the "early time" or semi-infinite solution, is given by

$$h_u/H \cong \text{EXP}(\alpha\beta^2)\ \text{ERFC}(\alpha\beta^2)^{1/2} \tag{4.17}$$

An approximate solution for large dimensionless times is given by (Hsieh et al., 1981):

$$\frac{h_u}{H} \cong \frac{1}{1+\beta+\gamma} + \frac{2\ \text{EXP}(-\alpha\phi_1^2)(\beta+\gamma^2\phi_1^2/\beta)}{\left[\gamma^2\frac{\phi_1^4}{\beta^2} + (\gamma^2\beta+\gamma^2+\gamma+\beta)\frac{\phi_1^2}{\beta} + (\beta^2+\gamma\beta+\beta)\right]} \tag{4.18}$$

and

$$\frac{h_d}{H} \cong \frac{1}{1+\beta+\gamma} + \frac{2\ \text{EXP}(-\alpha\phi_1^2)(\beta-\gamma\phi_1^2/\beta)}{\left[\gamma^2\frac{\phi_1^4}{\beta^2} + (\gamma^2\beta+\gamma^2+\gamma+\beta)\frac{\phi_1^2}{\beta} + (\beta^2+\gamma\beta+\beta)\right]\cos\phi_1} \tag{4.19}$$

ϕ_1 is the first root of equation 4.15.

When $\gamma \to 0$, or the downstream face of the sample acts an an impermeable boundary, the solution for dimensionless hydraulic head in the upstream reservoir becomes (Hsieh et al., 1981):

$$\frac{h_u}{H} = \frac{1}{1+\beta} + 2\sum_{m=1}^{\infty}\frac{\text{EXP}(-\alpha\phi_m^2)}{\phi_m^2/\beta+\beta+1} \tag{4.20}$$

where ϕ_m are the roots of

$$\tan\phi = -\phi/\beta \qquad (4.21)$$

When $\gamma \rightarrow \infty$, or the downstream reservoir is much greater than the upstream reservoir, the solution for dimensionless hydraulic head in the upstream reservoir is (Hsieh et al., 1980)

$$\frac{h_u}{H} = 2 \sum_{m=1}^{\infty} \frac{EXP(-\alpha\phi_m^2)}{\phi_m^2/\beta + \beta + 1} \qquad (4.22)$$

where ϕ_m are the roots of

$$\tan\phi = \beta/\phi \qquad (4.23)$$

4.4 Consolidation Test Theory

4.4.1 Introduction

The primary objective of bentonite consolidation testing is to determine the significance of consolidation on hydraulic conductivity. A secondary objective is to attempt to identify the outflow f action obtained due to consolidation drainage.

When a material consolidates, its void ratio decreases, thereby reducing the total pore spaces available for fluid flow. The hydraulic conductivity of the material should decrease with such a reduction. Other processes, such as increasing moisture content, may offset this reduction in hydraulic conductivity. This may apply to bentonite. Hamilton and Crawford (1959) report that the effect of consolidation on the void ratio may not be uniform throughout the material. The same may be true for the moisture content. Zones of changing hydraulic conductivity may develop because of the non-uniformity in water content and degree of compaction over time when bentonite is subjected to loading. This is particularly relevant for in-hole compacted clay for which the installation may be quite similar to that in a consolidation test ring. Tavenas et al. (1983) discuss various aspects of inhomogeneity implications of permeability testing in swelling and consolidating clays.

During constant pressure injection testing, the bentonite is subjected to loads or hydraulic heads for varying time periods. The clay undergoes some volume reduction or consolidation resulting from expulsion of water. Determining hydraulic conductivity values from a constant injection test requires knowledge of effluent volume flowing through the material over some time period. Consolidation contributes to the volume flowing out of the material, possibly resulting in high effluent volume measurements. Hydraulic conductivity values thus determined would be higher than those obtained from true flow through the undeformed material only.

4.4.2 Consolidation Theory

Consolidation is defined as the process whereby an initially saturated soil or a soil compacted to the point of saturation is compressed in a manner that results in reduction of pore volume by expulsion of water (Terzaghi, 1943, p. 265). Consolidation is not an instantaneous process, but rather one due to long term loading.

Terzaghi (1943, pp. 265-296) formulated a description of the consolidation process known today as the Terzaghi consolidation theory. It is based on several assumptions:

1) The soil is completely saturated.
2) The water and soil particles are incompressible.
3) Water flow and consolidation occurs in only one dimension.
4) Darcy's law is valid.
5) Hydraulic conductivity is constant.
6) Loading is instantaneous and constant, and the time lag of compression is caused entirely by the slow outflow of water from the compressing soil.
7) There is a linear relationship between the effective stress acting on the soil matrix and the volume change produced (Terzaghi, 1943, pp. 266-267).

Since the reduction in soil volume is assumed to be due to, and equal to, the change in void volume, a continuity equation of can be written:

$$\frac{\partial V_v}{\partial t} = -K \frac{\partial^2 h}{\partial z^2} \qquad (4.24)$$

where V_v = volume of voids
h = pressure head of soil water = $P/\rho g$ (actual pore pressure divided by liquid density and acceleration due to gravity)
z = coordinate in vertical direction.

The coefficient of volume change, M_v, relates the change in void volume to the change in pore pressure:

$$M_v = -\frac{\partial V_v / \partial t}{\partial P / \partial t} = -\frac{\partial V_v}{\partial P} \qquad (4.25)$$

Substituting Equation (4.25) into (4.26) yields

$$M_v = \frac{K}{\rho g} \frac{\partial^2 P / \partial z^2}{\partial P / \partial t} = \frac{\partial P}{\partial t} \qquad (4.26)$$

since $M_v = (\partial P)/(\partial t)$, Equation (4.26) can be written as

$$\frac{\partial P}{\partial t} = \frac{K}{\rho g M_v} \frac{\partial^2 P}{\partial z^2} = C_c \frac{\partial^2 P}{\partial z^2} \qquad (4.27)$$

where $C_c = K/(\rho g M_v)$ = coefficient of consolidation (Terzaghi, 1943, p. 271).

The rate of consolidation of the soil is determined by the hydraulic conductivity and by the coefficient of consolidation. These two properties are determined with laboratory tests, generally on small undisturbed cores placed in an oedometer. The sample is incrementally loaded and the deformation of the sample is monitored. The hydraulic conductivity of the sample is determined by either constant or falling head testing after each loading period. This type of testing has two problems for bentonite: the procedure is time consuming and may exceed realistic laboratory limits, and it creates a pressure gradient along the sample. Consolidation continues, resulting in questionable hydraulic conductivity values (Znidarcic, 1982, p. 90; Tavenas et al., 1983).

The consolidation test procedures assume that over a reasonable testing period the excess pore pressures induced by loading will dissipate (Holtz and Kovacs, 1981, pp. 286-291). The rate of dissipation is a function of the permeability. Terzaghi states that "changes in water content due to changes in the state of stress take place very slowly, because the low permeability of the soil does not permit a rapid transfer of water from one part of the soil mass to another" (Terzaghi, 1943, p. 265). For clays such as bentonite, consolidation may be too slow to measure.

CHAPTER FIVE

LABORATORY TESTING APPARATUS AND PROCEDURES

5.1 Introduction

The objective of this laboratory testing was to determine the effects permeants have on the hydraulic conductivity of bentonite. Constant pressure injection and transient pulse testing was conducted using permeants differing in chemical composition. Section 3.4 describes the permeants.

Distilled water, because of its low electrolyte content, was used to obtain a set of reference hydraulic conductivity values. These reference values were used to assess the performance of bentonite subjected to permeants having higher electrolyte concentrations. Groundwater obtained in northern Texas from the Ogallala aquifer was the primary permeant. Concentrations of major ions were determined to characterize the groundwater and possibly relate changes in measured hydraulic conductivity of the tested bentonite to a specific ion. The effects of sodium and calcium cations on the conductivity of bentonite were investigated using permeants with calcium and sodium concentrations similar to those of the groundwater. Finally, a relatively high concentration calcium water was injected into bentonite plugs in an effort to induce cation exchange of calcium for sodium adsorbed on the clay mineral surface.

Subjecting the plugs to permeants likely to occur in nature and to synthetic permeants designed to isolate the effect of a single ion provides important information about the permeability behavior of bentonite clay. An understanding of how bentonite behaves when subjected to permeants is important when considering bentonite for sealing applications.

Hydraulic conductivity testing was conducted in a temperature controlled environment. Temperature, measured daily throughout the test period, averaged 27 $\pm$ 2°C. Evaporation was monitored from pipets and burets similar to those used for effluent collection. Results of the evaporation studies are presented in Table 5.1. The 10 ml graduated cylinder had a surface area similar to that of the collection burets used in the Ogallala (Section 5.2.1) and low calcium concentration (Section 5.2.3) systems. The 10 ml pipet had a surface area similar to that of the collection pipets used in the distilled water system (Section 5.2.1) and the consolidation experiments (Section 5.4). The surface area of the 10 ml buret approximated the surface area of the collection pipets used in the high calcium (Section 5.2.2) and low sodium concentration (Section 5.2.3) systems. Evaporation was minimal. Therefore, no adjustment was made of the collection volumes used in Darcy's law.

Table 5.1 Evaporation Measured on Effluent Collection Devices

Collection Device	Average Measured Evaporation (ml)	Test Cycle (days)	Average Evaporation per Day (ml/day)
10 ml graduated cylinder	3.0	23	0.13
10 ml pipet	0.45	38	0.01
10 ml buret	0.85	38	0.02

With the exception of the bentonite plugs tested in the high calcium concentration system (Section 5.2.2), all bentonite plugs were constructed following American Colloid's recommendation, and a widely used hole sealing practice, of dropping Volclay tablets through water. Although void spaces, which can influence the hydraulic conductivity of the plug, were visibile along the clay and plug housing interface, no effort was made to compact the plug to a more homogeneous state.

5.2 Constant Pressure Testing Equipment and Procedures

5.2.1 Distilled Water and Ogallala Groundwater Systems

Constant pressure injection testing of bentonite was conducted using the apparatus illustrated in Figure 5.1. It consists of the nitrogen over water delivery assembly and the bentonite plug assembly. Combining the two provides a means of conducting constant pressure injection testing as well as transient pulse testing.

The nitrogen over water assembly delivers the permeant under a constant head to the bentonite plug assembly. The delivery system consists of two (2.5 cm) 1-inch i.d. polyvinylchloride (PVC) tubes 96.5 cm (38 inches) long. Each tube is capped with 2.51 cm (1 inch) diameter PVC I schedule 80 caps into which 6 x 6 mm (1/4 x 1/4 inch) swagelok fittings are inserted. 6 mm (1/4 inch) diameter copper tubing connects the water delivery system to the pressurized nitrogen source. Nitrogen is used because it is essentially an inert gas and has a relatively low water solubility. The CRC Handbook of Chemistry and Physics (1982, p. B125) gives the solubility of nitrogen in water at 1 atmosphere as 2.33 cc per 100 cc of water at 0°C and 1.42 cc per 100 cc of water at 40°C. Helium, with a water solubility of 0.94 cc per 100 cc of water at 0°C and 1.05 cc per 100 cc of water at 50°C (CRC Handbook of Chemistry and Physics, 1982, p. B104) may be more desirable. A valve between the nitrogen source and the water delivery system allows the reservoirs to be refilled without disturbing the nitrogen tank regulator. The plug assembly can be isolated from the water reservoirs

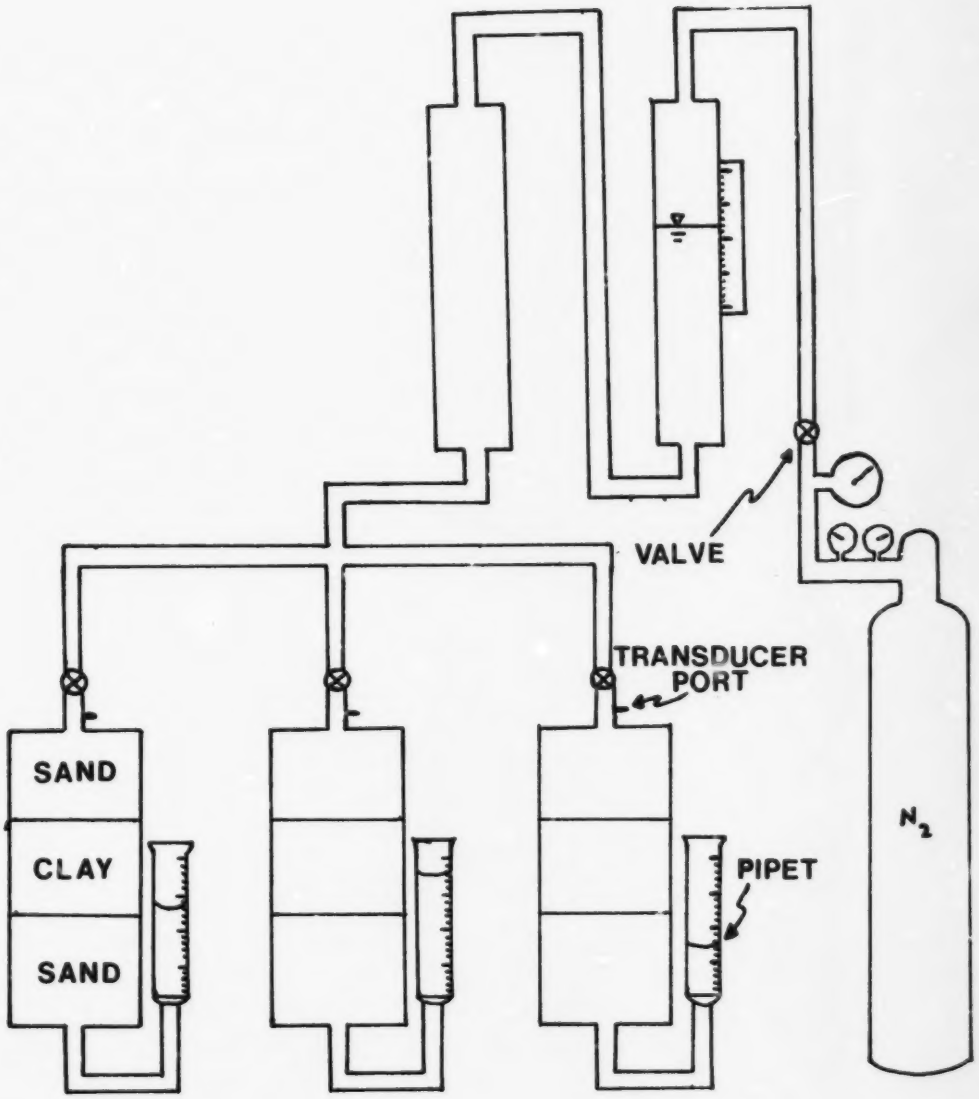

Figure 5.1 Schematic of apparatus used for constant pressure injection testing with distilled water and with Ogallala groundwater.

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by a valve between the reservoir and the transducer port. This prevents pressurization loss in the plug assembly when the reservoirs are opened for replenishing. All permeants are degassed before being introduced to the reservoirs. Degassing is accomplished by boiling the permeant for approximately 5 minutes and overfilling the reservoirs. Overfilling displaces air in the reservoirs. A scale with twenty divisions per inch (2.54 cm) mounted on the reservoir allows monitoring the flow into the plug assemblies.

The bentonite plug assembly consists of a 10.4 cm (4 inch) i.d. PVC tube 38.1 cm (15 in) long sealed at both ends with 10.4 cm (4 inch) diameter PVC I schedule 80 caps. One 6 x 6 mm (1/4 x 1/4 inch) swagelock fitting is inserted into each cap. 6 mm (1/4 inch) diameter copper tubing connects the bentonite plug assembly to the water delivery system. 6 mm (1/4 inch) diameter tygon tubing connects the plug assembly to the effluent collection device. Effluent was collected in 25 x 1/10 ml measuring pipets and 50 x 1/10 ml burets for the distilled water and groundwater systems, respectively.

The bentonite plugs utilized for distilled water testing were built over columns of saturated sand, each 1400 cm^3 in volume. The sand was washed with distilled water prior to plug construction. It was dropped into a column of distilled water. Each plug was constructed by dropping 250 1/2-inch (1.27 cm) diameter American Colloid Volclay bentonite tablets through 1100 ml of degassed, distilled water standing above the sand column. The tablets were allowed to hydrate and swell for 20 days before a confining layer of sand was added to each tube. The sand was dropped through a column of degassed, distilled water in an effort to prevent air from occupying pore spaces. At the end of the 20 day hydration/swelling period, the plugs had an average length of 16.0 cm (6.3 inches).

The bentonite plugs tested with Ogallala groundwater were constructed in the same fashion as those used in the distilled water testing except that groundwater from the Ogallala aquifer was substituted for distilled water in all phases of plug preparation. Groundwater degassing by boiling may have resulted in concentrating solutes in the groundwater slightly due to a small evaporation loss. The bentonite tablets were allowed a 20 day hydration/swelling period before adding the confining sand layer and sealing the PVC tubes. The final average plug length was 13.5 cm (5.3 inches), 2.5 cm (1 inch) shorter than the distilled water plugs. A possible explanation for this difference is offered in Section 6.2.2

Constant pressure injection testing is conducted using an initial pressure of 34 kPa (5 psi) for 30 days. Injection pressures are then increased to 69 kPa (10 psi) for 30 days, followed by a final 30-day period at 138 kPa (20 psi). Influent and effluent readings are made every 12 hours. Using Darcy's law, hydraulic conductivity values are calculated from the volume of effluent collected in a given time.

5.2.2 High Calcium Concentration System

Constant pressure injection testing of bentonite using a 1000 ppm calcium concentration permeant utilized the apparatus illustrated in Figure 5.2. It is similar to the one described in Section 5.2.1, but includes separate permeant reservoirs for each plug assembly. The bentonite plug construction method also differed.

Since clay plugs utilized in this phase of testing were constructed of bentonite having different initial moisture contents, separate permeant reservoirs were employed for each plug. Simply averaging the total volume injected over the three plugs was not felt to be reliable for determining the water intake of each plug.

The clay plugs were constructed from American Colloid tablets hydrated in the amount of water required to obtain a given moisture content (by weight). The plastic containers of water and clay were sealed and left undisturbed for 14 days. At that time the clays were inspected for homogeneity. Clays with moisture contents of 100 and 125 percent required some mixing to move unadsorbed water into contact with the clay. The containers were then resealed and left undisturbed for an additional 7 days. Clays with moisture contents of 50, 75, 100 and 125 percent (by weight) were prepared with distilled water and with Ogallala groundwater. Table 5.2 summarizes the moisture content of the clay, as determined by oven drying.

Clays prepared from distilled water and having moisture contents of 75, 100 and 125 percent were utilized in the high calcium concentration system. A 2.5 cm (1 inch) long mold with an inside diameter of 5.1 cm (2 inches) was used to form the plugs. Clay with a specific moisture content was added to the mold and gently formed into a 2.5 cm (1 inch) long plug. The plug was carefully positioned on top of a sand column comprising the large reservoir. This reservoir had been saturated with the 1000 ppm calcium permeant. The small reservoir was saturated with distilled water. A competent seal was formed between the PVC tube walls and the plug by gently smearing the clay plug against the tube walls. Throughout this operation, care was taken to avoid excessive compaction of the clay plug. Once in place, the confining sand layer was added and the tubes were sealed. A 10 to 1 ratio between the large and small reservoirs was maintained to comply with recommendations by Hsieh et al. (1981) for transient pulse testing. The permeant was degassed before being introduced to the permeant reservoirs.

Testing was conducted using an injection pressure of 17 kPa (2.5 psi). This pressure was used rather than higher injection pressures in an attempt to avoid puncturing the 2.5 cm (1 inch) long bentonite plug. Influent and effluent readings were made every 12 hours. An engineer scale, having twenty divisions per inch (2.54 cm), mounted on each reservoir, allowed measuring the flow into the plug assemblies. Hydraulic conductivity values were calculated using Darcy's law and the volume of effluent collected in a specified time. Effluent was collected in 10 x 1/10 ml burets.

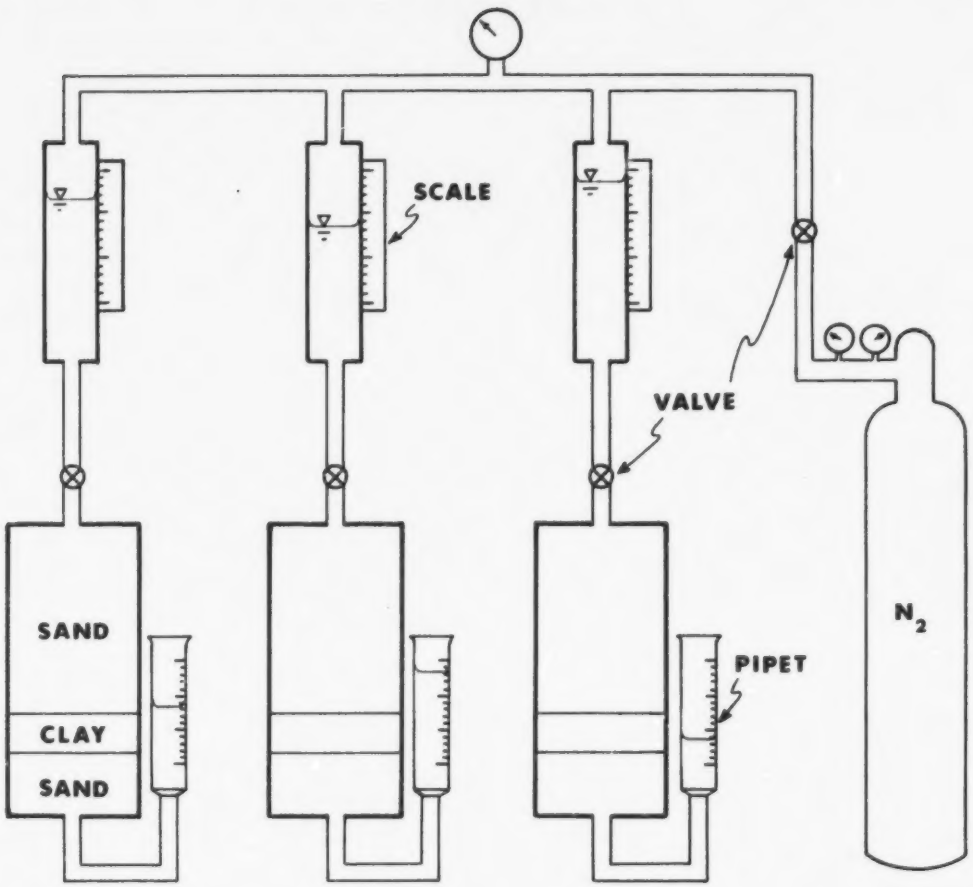

Figure 5.2 Schematic of constant pressure injection testing apparatus for water with high calcium concentration.

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Table 5.2 Moisture Content of Premixed Bentonite

Clay Hydrated With Distilled Water		Clay Hydrated With Ogallala Groundwater
Design Moisture Content (%)	Actual Moisture Content (%)	Actual Moisture Content (%)
50	54	53
75	78	84
100	99	95
125	123	127

5.2.3 Calcium and Sodium Systems

Constant pressure injection testing of bentonite using permeants representative of the calcium and sodium concentrations in the Ogallala groundwater utilized an apparatus similar to the one described in Section 5.2.1. The plug housing for both systems consists of PVC tubes with an inside diameter of 5.1 cm (2 inches) and a length of 41.7 cm (16.4 inches). Each tube is sealed with a PVC cap into which 3 x 6 mm (1/8 x 1/4 inch) swagelok fittings are inserted. The permeants include a 45 ppm calcium water made by dissolving calcium chloride in distilled water and a 39 ppm sodium water made by dissolving sodium chloride in distilled water. Both permeants underwent degassing before being introduced to the reservoirs.

The bentonite plugs were built over columns of saturated sand. Degassed, distilled water was used for washing and saturating the sand columns comprising the small reservoirs. The sand columns comprising the large reservoirs of the calcium system were saturated with the 45 ppm calcium permeant. Those of the sodium system were saturated with the 39 ppm sodium permeant. The sand columns were designed so that the ratio of the large reservoir to the small reservoir was 10 to 1. This was to comply with recommendations by Hsieh et al. (1981) in the event that transient pulse testing were to be performed on these plugs. Each plug was constructed by dropping 20 Volclay tablets through approximately 200 ml of degassed, distilled water. They were allowed to hydrate and swell for 20 days before adding the confining sand layer and sealing the tubes. It was necessary to remove approximately 2.5 cm (1 inch) of the plug after the hydration/swelling period ended so as to have a plug 2.5 cm (1 inch) in length. 10 x 1/10 ml measuring pipets were used to collect the effluent from the sodium system while 50 x 1/10 ml burets were used with the calcium system. The collection devices were connected to the PVC tubes by 6 mm (1/4 inch) diameter tygon tubing.

Injection pressure and monitoring frequency for the calcium and sodium systems were the same as for the high calcium system.

5.3 Transient Pulse Testing Equipment and Procedures

Transient pulse testing of bentonite plugs was conducted in two phases. In the first phase, transient pulse testing was performed on the bentonite plugs tested with the distilled water and with the Ogallala groundwater systems (Section 5.2.1). Transient pulse testing of these plugs was conducted prior to constant pressure injection testing. In the second phase, bentonite plugs constructed from clay with a predetermined moisture content were subjected to transient pulse testing.

Transient pulse testing of bentonite plugs utilized in the distilled water and Ogallala groundwater systems was accomplished using the apparatus illustrated in Figure 5.3. A nitrogen-over-water delivery system was used to introduce a pressure slug to each plug housing. Pressure decay in the upstream reservoirs was monitored by 34 and 340 kPa (5 and 50 psi) Dynisco transducers. Valves between the transducers and the water delivery systems isolate the bentonite plug assembly from the pressure source. The swagelok fitting in the lower endcap is capped, resulting in complete isolation of the plug assembly. The ratio of the compressive storage in the downstream reservoir to the compressive storage in the upstream reservoir was 0.87 and 0.76 for the distilled water and Ogallala groundwater systems, respectively. The difference in this ratio between the two systems is due to differences in swelling of the plugs (Sections 6.2.1 and 6.2.2).

Transient pulse testing of bentonite plugs constructed from clay having a predetermined moisture content was accomplished using an apparatus similar to that illustrated in Figure 5.3. A nitrogen-over-water delivery system was used to introduce a pressure slug to each plug housing. Pressure decay in the upstream reservoirs was monitored by 34 kPa (5 psi) Dynisco transducers, while 340 kPa (50 psi) Dynisco transducers monitored pressure buildup in the downstream reservoirs. During plug construction, an effort was made to maintain a 10 to 1 ratio between the downstream and upstream compressive storages. However, matching of the experimental curve to the theoretical type curve indicates that this ratio was not maintained during testing (Section 6.4.2). Reservoir compressive storage values calculated from experimental data are similar to those obtained from curve matching techniques. The actual ratio of the compressive storage of the downstream reservoir to the compressive storage of the upstream reservoir was 2.97 for the Ogallala groundwater system and 2.44 and 7.15 for the distilled water system.

The plug housing assembly consisted of a 38.1 cm (15 inch) long steel pipe having an inside diameter of 3.8 cm (1.5 inches). Threaded steel caps, fitted with 3 x 3 mm (1/8 x 1/8 inch) swagelok fittings, were used to seal each end of the pipe. 3 mm (1/8 inch) I.D. nylon tubing connected the plug housing with the permeant reservoir. A valve between the transducer and the permeant reservoir isolated the plug housing from the pressure source. 6 mm (1/4 inch) I.D. nylon tubing

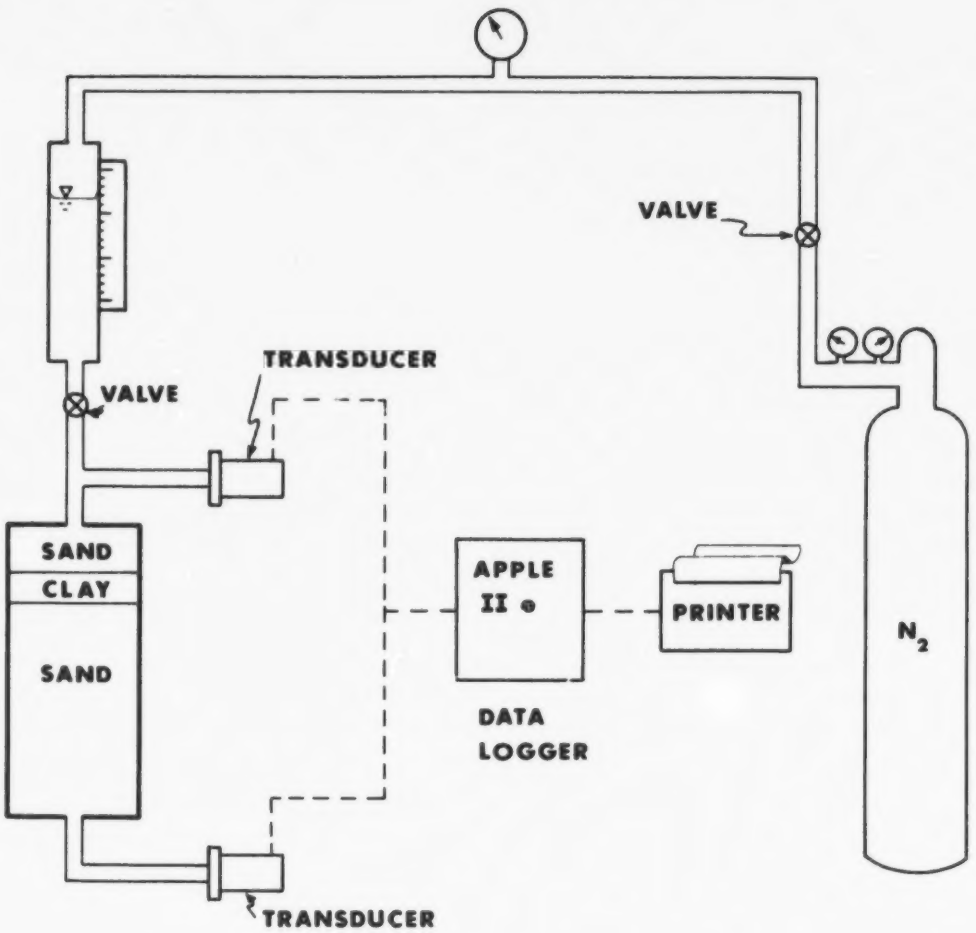

Figure 5.3 Schematic of apparatus used in transient pulse testing of bentonite plugs.

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connected the permeant reservoir to the pressure source. A 138 kPa (20 psi) gage, connected to the pressure injection line, monitored the injection pressure. A valve between the gage and pressure source prevented the pressure pulse from affecting the permeant reservoirs until the desired pressure was obtained.

Clays prepared from American Colloid Volclay tablets and hydrated with sufficient distilled water or Ogallala groundwater to attain moisture contents of 75, 100 and 125 percent (by weight) were used in the second phase of transient pulse testing. With one exception, plug construction was accomplished as described in Section 5.2.2. The bentonite plugs were 1 cm (0.4 inch) long and 3.8 cm (1.5 inches) in diameter.

Transient testing was accomplished by introducing a pressure slug to the upstream reservoir. Transducers connected to the upstream and downstream reservoirs and to a multi-channel data-logger monitored pressure changes in the reservoirs. The data-logger converts the transducer signals for the Apple II-E microcomputer, which outputs the reservoir pressures at 20-minute intervals.

5.4 Consolidation Test Equipment and Procedure

Consolidation testing of bentonite plugs was accomplished using an apparatus similar to that used for constant pressure testing, Figure 5.1. No water reservoir is required as the pressure is introduced directly from the nitrogen source to the plug assembly. The plug housing consists of PVC tubes having an inside diameter of 10.16 cm (4 inches) and a length of 30.5 cm (12 inches). The clay plug was constructed by dropping 250 American Colloid Volclay tablets through a column of degassed Ogallala groundwater standing on a column of sand. The height of the sand column was reduced to accommodate the shortened PVC tube serving as the plug housing. The tablets were allowed to hydrate and swell for 20 days before the PVC tubes were sealed. No confining sand column was added. The final plug heights were measured. Collection of outflow was accomplished using three 25 x 1/10-ml pipets. A gage located between the plug assembly and pressure source monitored the injection pressure.

Consolidation pressure was initially set at 34 kPa (5 psi), and subsequently increased to 69 and 138 kPa (10 and 20 psi). Early time outflow measurements were made when a pressure increment was applied, at 30 seconds, one minute, two minutes, four minutes, eight minutes and 16 minutes. The duration of testing at a given pressure was governed by the duration of constant pressure testing at the same pressure.

Dehydration of the plug surfaces due to the use of nitrogen as a pressure source induced cracks in the plugs. They deepened and widened with time. When testing at a given pressure was completed, the plug assemblies were opened and 80 to 100 ml of Ogallala groundwater was added to each tube. After 24 to 48 hours, no standing water was observed above any of the plugs, and the cracks had healed. The tubes were then closed and testing resumed.

CHAPTER SIX

TEST RESULTS AND DISCUSSION

6.1 Introduction

The primary objective of this testing is to determine the effects of permeant chemistry and composition on the ability of bentonite to transmit water. Processes that significantly influence the hydraulic conductivity of bentonite include consolidation, swelling, dispersion, and flocculation. All four are affected by permeant chemistry.

Consolidation is time-related, involving compression, stress transfer, and drainage. A soil with slow drainage, such as bentonite, requires a long time to consolidate. Processes which change drainage characteristics influence consolidation. Therefore, factors which affect the hydraulic conductivity, such as the diffuse double layer, influence consolidation.

Swelling is affected by permeant chemistry. The ratio of sodium to calcium cations in the permeant and in the clay structure, as well as the total salt concentration of the permeant, affect the affinity of the clay mineral for water and hence, its swelling pressure. The thickness of the adsorbed water surrounding a clay particle may increase or decrease, depending upon the chemical makeup of clay and permeant.

Clay particles carry negative charges and will not attract or collide in distilled water. The negative charge causes interparticle repulsion, and particles may remain in suspension for some time due to Brownian motion. They remain dispersed until an electrolyte containing positively charged ions or colloids is added to the suspension. The positively charged ions or colloids are attracted to the negatively charged clay particles, resulting in formation of larger colloidal clumps or flocs. This process is called flocculation. A well dispersed clay will generally be less permeable than the same clay in a flocculated state.

6.2 Constant Pressure Injection Testing

Constant pressure injection testing was conducted on bentonite plugs constructed using distilled water and Ogallala groundwater, as well as on bentonite plugs with a predetermined moisture content. Permeants of different compositions were used. Sections 6.2.1 and 6.2.2 present the results of constant pressure injection testing of the distilled water and Ogallala groundwater systems. Section 6.2.3 presents the results of constant pressure injection testing on bentonite plugs with predetermined moisture contents using a high calcium concentration permeant. Sections 6.2.4 and 6.2.5 present results of constant injection testing of bentonite plugs using permeants with low calcium

and sodium concentrations. Section 6.4.2 presents results of transient pulse testing of 1 cm (0.39 inch) long bentonite plugs constructed using distilled water or Ogallala groundwater.

6.2.1 Distilled Water System

The theory, equipment and procedures used in constant pressure injection testing have been described in Sections 4.2 and 5.2. Three bentonite plugs, hydrated with distilled water, were tested using distilled water under pressures of 34, 69 and 138 kPa (5, 10, and 20 psi). Approximately one month was allowed for testing at each pressure.

The clay tablets were allowed to hydrate and swell for 20 days before adding the confining sand layer and sealing the plug housing. This ensured the clay had sufficient time to expand to its free swell capacity. Free swell is defined as (Holtz and Kovacs, 1981, p. 189):

$$[(\text{FINAL VOLUME} - \text{INITIAL VOLUME})/\text{INITIAL VOLUME}] \times 100 \qquad (6.1)$$

and is expressed as a percent. After approximately 16 days, the free swell remained constant. The final average plug height was 16.0 cm (6.5 inches).

Hydraulic conductivities calculated from measured outflow average approximately 10^{-8} cm/s for all three plugs. Figures A.1 through A.12 (Appendix A) show the change in hydraulic conductivity with time. Table 6.1 summarizes the results. The hydraulic conductivities were calculated assuming that all flow from the plugs is flow through the plugs. A portion of this flow may be due to consolidation. An adjustment to the measured outflow would therefore appear to be in order. The outflow volume, adjusted for drainage due to consolidation, would be used to calculate the hydraulic conductivity. An independent experiment, discussed in Section 6.3, was conducted to determine the effluent volume due to consolidation drainage. This approach met with very limited success, as did similar trials by Kimbrell et al. (1987). Consequently, all hydraulic conductivity values derived from constant pressure injection testing reported in this and in subsequent sections use the total measured outflow rather than an adjusted volume. The actual hydraulic conductivity values may therefore be smaller than reported.

Figures A.1 through A.12 (Appendix A) show relatively high hydraulic conductivity values for the first day of testing at each pressure increment. This peak may in part be due to increased consolidation drainage resulting from an increase in the applied pressure. The mass balance plots shown in Figures B.1 through B.12 (Appendix B) appear to support this. The plots show large volumes on the first day of testing at each pressure. These peaks appear on the second day of testing at 34 and 138 kPa (5 and 20 psi). Testing at 69 kPa (10 psi) began ten to twelve hours earlier than testing at 34 and at 138 kpa. Had the outflow volume been combined with the second day's volume, the large outflow peaks would shift to the first day. The hydraulic conductivity values are not affected since time is an integral part of Darcy's law.

Table 6.1 Average Hydraulic Conductivity Results. Bentonite Plugs Tested with Distilled Water. Constant pressure injection.

Pressure kPa (psi)	Hydraulic Conductivity (cm/s) Tube #1	Tube #2	Tube #3	Total System
34 (5)	2.0×10^{-8}	1.8×10^{-8}	1.9×10^{-8}	1.9×10^{-8}
69 (10)	1.6×10^{-8}	1.5×10^{-8}	1.6×10^{-8}	1.5×10^{-8}
138 (20)	1.0×10^{-8}	9.8×10^{-9}	1.0×10^{-8}	1.0×10^{-8}

The average daily outflow volumes and hence the hydraulic conductivity remain approximately constant for each pressure increment following the occurrence of these peaks.

Examination of the mass balance plots suggests that the clay experiences significant changes in moisture content during the initial testing period at each pressure increment. These changes depend on experimental conditions. Following initial hydration of the bentonite tablets, the plug is sandwiched between two saturated sand columns. The plug housing is then sealed, thereby restricting the plug volume. Pressure applied to a clay under these conditions initiates consolidation. Consolidation reduces the volume of voids. The volume reduction allows the clay minerals to adsorb additional water until the clay returns to its original volume. A new steady state or equilibrium state is thus achieved. At this point, the differences between influent and effluent volumes should be minimal.

The mass balance plots show relatively large injection volumes at the beginning of each pressure increment. Some of this may be attributable to water compressibility and compressibility of the apparatus. This instantaneous compression would have little effect beyond the time of the initial pressure increase. The plots also show significantly reduced effluent volumes with respect to injection volumes. A considerable difference between influent and effluent volumes exists in the initial stages of testing. As the moisture content increases and a new equilibrium condition is approached, the difference decreases. Approximately 13 days are required for a new equilibrium condition to be established at 34 kPa (5 psi). Three to four days are required at 69 and 138 kPa (10 and 20 psi). Unusually high influent peaks appear on day 25 and 17 for testing at 69 and 138 kPa (10 and 20 psi), respectively. An inspection of the daily record shows that on day 25, 10.3 ml of water was injected into the 69 kPa (10 psi) system over 497 minutes. On day 17, 20.3 ml was injected into the 138 kPa (20 psi) system over 414 minutes. Normally, 4 to 6 ml would be expected to be injected over comparable periods. Further inspection of the record shows that the ruler used to measure changes in the reservoir water level was repositioned on both days. This suggests that the ruler was not securely fastened to the reservoir, resulting

Table 6.2 Average Hydraulic Conductivity during the Final 16 Days of Each Pressure Increment of Bentonite Plug Testing Using Distilled Water

Pressure kPa (psi)	Hydraulic Conductivity (cm/s) Tube #1	Tube #2	Tube #3	Total System
34 (5)	1.6×10^{-8}	1.6×10^{-8}	1.7×10^{-8}	1.6×10^{-8}
69 (10)	1.3×10^{-8}	1.2×10^{-8}	1.3×10^{-8}	1.3×10^{-8}
138 (20)	8.0×10^{-9}	7.8×10^{-9}	7.9×10^{-9}	7.9×10^{-9}

in some slippage. After a new reference position was established, influent volumes returned to normal.

Hydraulic conductivity values in Table 6.1 show a decrease in conductivity with increasing pressure. This suggests that pore volumes may be reduced by swelling and consolidation.

Hydraulic conductivity values in Table 6.2 represent averages of the last 16 days of constant pressure injection testing at each pressure. These are slightly lower than those in Table 6.1 and may be more representative. The systematic reduction in hydraulic conductivity with pressure suggests that swelling and consolidation may be occurring.

The hydraulic conductivity values can be compared with published results. Pusch et al. (1983) report a hydraulic conductivity of 10^{-10} to 10^{-12} cm/s for intensely compacted sodium-rich bentonites using constant head injection testing. Singh (1982) reports conductivities of 10^{-9} cm/s for similar clays. Hodges et al. (1982) report hydraulic conductivities ranging from 10^{-11} to 10^{-13} for similar clays using constant head injection testing. The hydraulic conductivity values reported in Table 6.1 are above the upper end of the range of those reported in the literature. If allowances are made to account for consolidation drainage, the conductivity values reported in this study should decrease. The plugs as tested here have not been compacted.

The final moisture content of each plug was determined at the completion of constant pressure injection testing. Three samples were collected from each plug and were oven-dried for 60 hours at 66°C (150°F). The samples were weighed after 48 hours, returned to the oven, and reweighed 12 hours later. If no significant weight changes were observed the sample was assumed to be completely dried. Samples were obtained from the upper and lower clay-sand interfaces and from the midsection of the plug. The string of sampling points was defined by a cylinder with a diameter of approximately 2.5 cm (one inch)

Table 6.3 Bentonite Plug Moisture Contents. Moisture Content at Three Locations of Each Distilled Water Bentonite Plug After Constant Pressure Injection Testing.

Sample Location	Moisture Content (%) Tube #1	Tube #2	Tube #3
Upper Interface	616	675	499
Midsection	279	230	202
Lower Interface	152	145	130

centered on the vertical axis of the plug. This axis was chosen for sampling because the plug-wall interface may not be representative, especially after four months of testing. Sampling was accomplished by inserting and rotating a 1.3 cm (0.5 inch) wide spatula through the length of the plug. This core was then sampled at the appropriate locations.

Table 6.3 summarizes the final moisture content of the three plugs. A moisture gradient is observed in each plug. The moisture content at the upper clay-sand interface ranged from 499% in tube 3 to 675% in tube 2. The median value was 597%. The moisture content reported at this interface for tube 3 may be in error. The moisture condition at this interface made sampling difficult. Upon removal of the top sand from the plug housing, a very fluid layer of clay approximately 3.8 cm (1.5 inches) deep was observed. The upper 1.3 cm (0.5 inch) of this layer had mixed with the sand column, forming a viscous gel. The gel did not flow after the sand column had been removed, but retained its shape and position in the plug housing, even after the housing had been placed on its side. When the clay-sand gel was removed, the remaining layer of very fluid clay flowed from the face of the more cohesive plug. While attempting to sample the fluid-like clay, care had to be taken to ensure that no sand grains were introduced into the sample. Since sampling was restricted to a cylinder centered about the vertical axis and extending through the plug length, the sample obtained from plug #3 may not have been representative. It is possible that the sample contained more of the cohesive portion of the plug than of the fluidized clay, resulting in a relatively lower moisture content compared to the other two plugs.

Sampling at the midsection and lower clay-sand interfaces presented no problems. Care in sampling at the lower interface was necessary to ensure that no sand grains were collected. Observed moisture contents at the midsections of the plugs ranged from 202% in tube 3 to 229 % in tube 1. The median value was 237%. The lower clay-sand interface exhibited moisture contents ranging from 130% in tube 3 to 152% in tube 1, with an average of 142%.

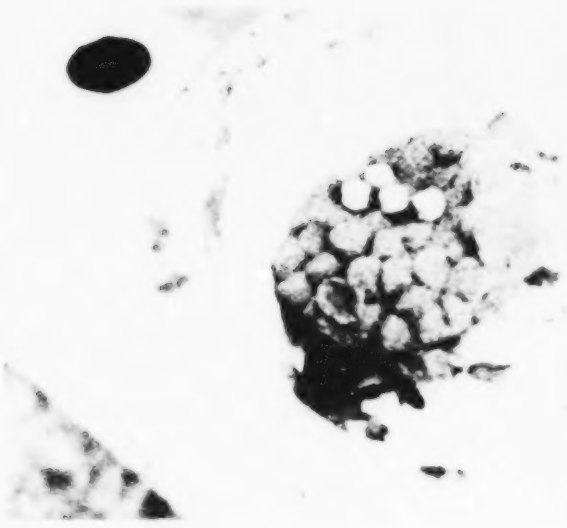

Figure 6.1 Partially swelled bentonite tablets which make up one of the plugs used in constant pressure injection testing. The plug was removed after about four months of flow testing. Four untested bentonite tablets are placed on top of the recovered plug section to allow a comparison between hydrated and fresh tablets.

Complete swelling of bentonite tablets is assumed to occur when they are immersed in water. Incomplete swelling of tablets used to form a seal might increase the ability of the seal to transmit water. Incomplete tablet swelling was observed in bentonite recovered from one of the tubes used in constant injection testing with the distilled water and Ogallala groundwater systems. Unfortunately, the specific tube could not be identified. The remnants of the plug containing the partially swelled tablets were not observed until all six plugs had been sampled and completely removed from the PVC tubes. The partially swelled tablets had the same shape as the dry tablets, as can clearly be seen in Figure 6.1, which shows partially swelled tablets making up part of one of the plugs. The partially swelled tablets can be compared to dry as-delivered tablets included in the photograph.

Nonuniform plug formation can have a significant effect on the hydraulic conductivity. Voids caused by nonuniform hydration and swelling may contain water, air or a combination of both. Water-filled voids could serve as fluid reservoirs, promoting further hydration and swelling of the tablets. The additional swelling would decrease the volume of the voids and, consequently, the hydraulic conductivity. Air-filled voids would restrict the flow of water through the plug, resulting in decreased hydraulic conductivity. Voids containing both water and air would be expected to promote swelling and restrict water transmission.

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Voids, presumably resulting from nonuniform tablet hydration and swelling, were observed along the plug-housing interface in all the plugs. It is uncertain whether voids were present in the interior of the plugs. When the plugs were sampled, no evidence of voids was found; however, the sampling technique may have closed voids.

6.2.2 Ogallala Groundwater System

Three bentonite plugs, hydrated with water from the Ogallala aquifer in northern Texas, were tested using an Ogallala groundwater permeant under pressures of 34, 69 and 138 kPa (5, 10 and 20 psi). Approximately one month was allowed for testing at each pressure.

The clay tablets were allowed to hydrate and swell as described in Section 6.2.1. The time required to achieve an unchanging free swell in the Ogallala groundwater system was approximately five days less than that required for the distilled water system. However, the confining sand layer was not added until the 20th day. The final average plug height was 13.5 cm (5.3 inches). This is 2.5 cm (1 inch) shorter than plugs constructed using distilled water.

Swelling in montmorillonites is essentially a two-step process. In the first step, interlayer or intracrystalline swelling, the montmorillonite takes up 2 to 4 monolayers of water between the unit layers, resulting in, at most, a doubling of the clay volume. Swelling continues with the clay adsorbing an amount of water many times the volume of the original clay. The additional swelling, a result of the double-layer repulsion between the surfaces of the individual clay particles, is known as osmotic swelling. The water tends to equalize the high concentrations of ions between two particles, which are so close together that their double layers overlap, and the low concentration of ions far away from the particle surfaces in the bulk solution (van Olphen, 1963, pp. 147-152).

The Ogallala groundwater, having a higher ion concentration than the distilled water, compresses the double layer, thereby reducing the range at which particles are repelled. This causes the reduction in swelling observed for the Ogallala groundwater plugs as compared to the distilled water plugs.

An experiment was conducted to determine the effect distilled water and Ogallala groundwater have on the free swell of the bentonite tablets used in this study. The test apparatus consisted of two 250 ml graduated cylinders. One cylinder contained 170 ml of distilled water. The other contained 170 ml of Ogallala groundwater. Twenty bentonite tablets were added to each cylinder. The initial volume of the tablets was noted and the cylinders were sealed with Parafilm to prevent evaporation. The initial tablet volume was 57 ml. In 20 days the tablets hydrated with distilled water expanded to 120 ml, the tablets hydrated with the groundwater to 95 ml. The free swell was calculated using Equation (6.1). The distilled water tablets had a free swell of 111%, the Ogallala groundwater tablets only 67%.

Table 6.4 Average Hydraulic Conductivity of Bentonite Plugs Tested with Ogallala Groundwater at Constant Injection Pressures

Injection Pressure kPa (psi)	Hydraulic Conductivity (cm/s Tube #1	Tube #2	Tube #3	System
34 (5)	7.3×10^{-9}	8.9×10^{-9}	9.2×10^{-9}	8.5×10^{-9}
69 (10)	7.8×10^{-9}	8.4×10^{-9}	9.0×10^{-9}	8.4×10^{-9}
138 (20)	5.2×10^{-9}	6.0×10^{-9}	6.4×10^{-9}	5.8×10^{-9}

Hydraulic conductivities calculated from measured total outflow average approximately 8×10^{-9} cm/s for all three plugs. Figures A.1 through A.12 (Appendix A) show the hydraulic conductivity over time. Table 6.4 summarizes the results. The hydraulic conductivity values are based on unadjusted flow volumes. The actual hydraulic conductivities values may be smaller. Table 6.5 summarizes the results for the last 16 days of testing. These are slightly lower than those in Table 6.4 and may be more representative of the system after an equilibrium condition has been established.

The hydraulic conductivity plots for the Ogallala system show similar conductivity peaks at the beginning of each pressure increment application as observed for the distilled water system. These peaks may be due to relatively high consolidation drainage. The mass balance plots shown in Figures B.13 through B.24 (Appendix B) seem to support this. A relatively large difference exists between the peaks initially, but within two to three days it is minimized, suggesting that a new equilibrium condition has been attained.

The measured outflow volumes and calculated hydraulic conductivities for the Ogallala system are smaller than those for the distilled water system. This appears to contradict what is expected, based on diffuse double layer theory and flocculation properties.

Table 6.5 Average Hydraulic Conductivity during Final 16 Days of Bentonite Plug Testing Using Ogallala Groundwater

Injection Pressure kPa (psi)	Hydraulic Conductivity (cm/s) Tube #1	Tube #2	Tube #3	System
34 (5)	6.5×10^{-9}	6.6×10^{-9}	7.5×10^{-9}	7.1×10^{-9}
69 (10)	6.4×10^{-9}	6.7×10^{-9}	7.9×10^{-9}	7.0×10^{-9}
138 (20)	5.2×10^{-9}	4.7×10^{-9}	5.2×10^{-9}	5.0×10^{-9}

The clay tablets were hydrated with a solution having a high total salt content compared to distilled water. The sodium and calcium concentrations were approximately 1.7 meq/l and 2.2 meq/l, respectively. Total solution concentration was 12 meq/l. Under these conditions, the diffuse double layer should be compressed, resulting in larger pore volumes. The solution also would tend to cause more flocculation, again resulting in larger pore volumes. It is possible that suspended matter in the unfiltered groundwater lodged in the clay pore spaces when the tablets were hydrated, thereby blocking the water conducting channels in the clay. Additional particles may have been introduced by the permeant during the testing. Because of the particle settling in the reservoirs, this addition should have been minimal. The particle count distribution conducted on both the distilled water and the Ogallala groundwater supports channel blockage. Channel blockage by colloidal matter could effectively reduce the hydraulic conductivity, thereby explaining the apparent discrepancy in conductivity between the distilled and groundwater systems.

The hydraulic conductivity results in Table 6.4 show nearly identical values for test pressures of 34 and 69 kPa (5 and 10 psi). At 138 kPa (20 psi) the hydraulic conductivity decreased approximately 30 percent. This may be due to a process proposed by Rowell et al. (1969). They showed that the solution concentration at which clay disperses depends on the applied mechanical stress. Application of sufficient mechanical stress to a clay can cause dispersion at solution concentrations not normally associated with dispersion.

Another possible explanation for the observed hydraulic conductivity behavior of the distilled water and the Ogallala groundwater systems is based on the original plug heights of each system. The average plug height for the Ogallala groundwater system was 2.5 cm (1 inch) shorter than for the distilled water system. The total plug volume for the Ogallala system was less than that for the distilled system, suggesting that the volume of voids may be smaller in the Ogallala system. Consolidation in the groundwater plugs at 34 and 69 kPa (5 and 10 psi) may not have been as significant as in the distilled water plugs. When 138 kpa (20 psi) was applied, the hydraulic conductivity in the groundwater plugs decreased, suggesting that significant consolidation may have occurred.

It is likely that a combination of at least these three mechanisms is responsible for the difference in the hydraulic conductivity between the distilled water and Ogallala groundwater systems.

The effluent curve for plug 1 at 138 kPa (20 psi) in Figure B.21 (Appendix B) is unusual compared to effluent curves for other plugs. It shows decreasing outflow beginning on day 3, and continuing until day 11. The outflow increases until day 15, at which time a decreasing trend starts. On day 19 this trend reverses, and on day 20 a maximum outflow volume of 13.4 cm^3 is measured. A decreasing trend then continues until day 28. Beginning on day 28, the influent and effluent curves closely parallel each other. No calculation errors were found. The relatively slow decreasing trend of the effluent curve followed by

Table 6.6 Bentonite Plug Moisture Contents. Moisture Content at Three Depths of Each Ogallala Groundwater Bentonite Plug After Constant Pressure Injection Testing

Sample Location	Moisture Content (%) Tube #1	Tube #2	Tube #3
Upper Interface	398	476	397
Midsection	165	163	149
Lower Interface	105	127	103

a sudden large increase in outflow volume suggests that some type of restriction may have developed either in the clay plug or in the collection device, causing a pressure buildup. When the pressure became sufficiently large, the restriction was cleared and an equilibrium condition similar to that observed in tubes 2 and 3 was established. The blockage may have been due to clay particle dispersion and migration, as described by Rowell et al. (1969) and Hunt et al. (1985).

The moisture content of each plug was determined at the completion of flow testing. Sampling was conducted as described in Section 6.2.2. Table 6.6 summarizes the final moisture contents. A moisture gradient is present in each plug. The moisture content at the upper clay-sand interface ranged from 397% in plug 3 to 476% in plug 2. The average value was 424%. A fluid layer similar to the one described in Section 6.2.2 was observed at the upper clay-sand interface of the Ogallala groundwater plugs. Upon removal of the confining sand, a fluid clay layer approximately 2.5 cm (1 inch) deep was exposed. The upper half of this layer had mixed with the sand column. This mixture of clay and sand retained its shape and position in the plug housing after the sand column had been removed and the plug housing was placed on its side. When the clay-sand mixture was removed, the remaining fluidized clay flowed from the more cohesive plug.

The moisture contents at the midsections of the plugs ranged from 149% in plug 3 to 165% in plug 1. The average value was 159%. The moisture content at the lower clay-sand interface ranged from 103% in plug 3 to 127 % in plug 2. The average value was 112%.

Major differences between the Ogallala groundwater and distilled water systems included reduced swelling, moisture content, and hydraulic conductivity. A fourth difference was observed, or more precisely, smelled. Upon removing the end caps from the plug housing, a very strong "r ten egg" smell, indicative of hydrogen sulfide, permeated the laboratory. Each plug contributed to the smell. The hydrogen sulfide may have formed from the sulfate in the groundwater. In the presence of certain bacteria, sulfate may be reduced to hydrogen sulfide. The bacteria use the oxygen tied up in the sulfate ligand

Table 6.7 Average Hydraulic Conductivity of Bentonite Plugs Using a 1000 ppm Calcium Permeant

Initial Moisture Content (%)	Average Hydraulic Conductivity (cm/s)
75	6.35×10^{-9}
100	9.17×10^{-9}
125	1.3×10^{-8}

when free oxygen or oxygen associated with nitrate is not available (Montgomery, 1985, p. 22). This observation indicates that bacterial or organic actions may have occurred, and may have to be considered for bentonite plug performance. These effects, not investigated here in any more detail, also may be responsible for the unexpectedly low hydraulic conductivity for bentonite tested with Ogallala water, as compared to distilled water.

6.2.3 High Calcium Concentration System

Three bentonite plugs with different initial moisture contents were tested over a 44-day period using a 1000 ppm calcium permeant pressurized at approximately 17 kPa (2.5 psi). The plugs were tested using the apparatus and bentonite plug construction of a given moisture content described in Section 5.2.2.

Figures A.13 through A.15 (Appendix A) show the hydraulic conductivity over time for each plug. Table 6.7 summarizes the results. The hydraulic conductivity values were calculated from unadjusted outflow

Table 6.8 Average Hydraulic Conductivity during the Final 16 Days of Bentonite Plug Testing with a 1000 ppm Calcium Permeant

Initial Moisture Content (%)	Average Hydraulic Conductivity (cm/s)
75	4.2×10^{-9}
100	5.9×10^{-9}
125	1.2×10^{-8}

volumes. The actual hydraulic conductivity values are likely to be smaller than those reported. Table 6.8 summarizes the results for the last 16 days of the test period. These may be more representative because they do not include the high initial effluent volumes attributed to plug consolidation.

With the exception of the 125% plug, relatively high hydraulic conductivity values were observed on the first day of testing. The plug housing containing the 125% moisture content plug had a leak around the swagelok fitting in the lower end cap. This tube was pressurized, causing the clay to consolidate, but no outflow was measured because of the leak. This tube was isolated from the system, repaired, and put back on-line seven days later. Conductivity, influent and effluent peaks were observed when the system was brought back on-line. However, they are not as large as those observed from the other two tubes. The plug housing containing the 75% moisture content plug also experienced a leak. This leak, around the swagelok fitting in the upper end cap, was much smaller than the one in the 125% plug assembly. This tube was off-line from days 3 through 7. When it was brought back on-line, relatively large conductivity, influent and effluent peaks were observed.

The mass balance plots (Figures B.25 through B.27 (Appendix B)) show relatively constant inflows and outflows for all three plugs . The 125% plug shows a large influent peak on day 27, very similar to the peak for the Ogallala groundwater system at 138 kPa (20 psi) shown in Figure B.21. The two peaks may have similar causes. The mass balance plots show minimal differences between the influent and effluent curves after the first two to three days of testing. This suggests that an equilibrium state has been achieved for the testing pressure.

A relationship between moisture content and hydraulic conductivity is apparent from Tables 6.7 and 6.8. The hydraulic conductivity is smallest for the 75% plug and greatest for the 125% plug. According to Lambe and Whitman (1969, p. 292), the degree of saturation of a soil has an important influence on its permeability: the higher the degree of saturation, the higher the permeability. The data in Tables 6.7 and 6.8 suggest that the apparent relationship between the degree of saturation and hydraulic conductivity could be extended to include a relationship between moisture content and hydraulic conductivity.

The final moisture content of the bentonite plugs was determined as described in Section 6.2.1. Table 6.9 summarizes the results. A moisture gradient is present in plugs 2 and 3. Plug 1 has a uniform moisture content. A relationship between moisture content and hydraulic conductivity was observed for each plug. However, the primary objective, initiating cation exchange, may not have been achieved. Had sodium ions adsorbed on the clay mineral surfaces been replaced by calcium, the hydraulic conductivity should have increased. The volume of permeant injected into each plug housing was insufficient to displace the distilled water originally occupying the pore spaces within the bentonite plugs and collection reservoirs.

Table 6.9 Bentonite Plug Moisture Content. Moisture Content at Three Depths of Each Bentonite Plug After Constant Pressure Injection Testing with 1000 ppm Calcium Water

Sample Location	Moisture Content (%) Tube #1	Tube #2	Tube #3
Upper Interface	126	143	218
Midsection	125	134	188
Lower Interface	128	130	171

Moisture content at the start of flow testing is given in Table 6.7.

An approximate pore volume of 21 cc was calculated for the bentonite plug using a plug volume of 53 cc and an estimated porosity of 40 percent. Davis (1969) gives typical porosity values for clay ranging from 40 to 70 percent. The volume of the collection reservoir was 25 cc. The cumulative effluent collected from the plugs having moisture contents of 75, 100 and 125 percent, was 26, 44 and 57 ml, respectively. With the exception of the plug having a moisture content of 125 percent, the volume of effluent is not sufficient to replace entirely the original pore water within each plug. It is also unlikely that the 1.2 pore volumes collected from the bentonite plug having a moisture content of 125 percent was adequate to achieve complete cation exchange. Increasing the test period would be necessary for significant cation exchange to occur under the given test conditions. Increasing the injection pressure may shorten the test period; however, the probability of puncturing the bentonite plugs would increase, and the hydraulic gradient could become totally unrepresentative.

A chemical analysis of the influent and effluent near the end of the test period showed similar calcium concentrations in both, suggesting that either cation exchange or channeling may have occurred. However, the small injection volumes and the relatively constant hydraulic conductivity do not support these possibilities. Contamination of the sample during the chemical analysis is a more probable explanation.

6.2.4 Low Calcium Concentration System

Three bentonite plugs were tested with a 45 ppm calcium permeant over a 55-day period under a 17 kPa (2.5 psi) pressure. Plug construction, experimental procedures, and test apparatus are described in Section 5.2.3.

Hydraulic conductivities calculated from measured outflow average approximately 10^{-8} cm/s. Figures A.16 through A.19 (Appendix A) show the change of hydraulic conductivity over time. Table 6.10 summarizes

Table 6.10 Average Hydraulic Conductivity of Bentonite Plugs Using a 45 ppm Calcium Permeant Water. Results obtained prior to (A) and after (B) repair of Tube #1.

Tube #	Hydraulic Conductivity (cm/s)	
	(A)	(B)
1	1.4×10^{-8} *	1.7×10^{-8}
2	2.0×10^{-8}	2.0×10^{-8}
3	1.4×10^{-8}	1.4×10^{-8}
System	1.6×10^{-8} *	1.7×10^{-8}

* Slightly undermeasured due to Tube #1 leak.

the results. It also shows the average hydraulic conductivity of the three plugs over the testing period. Hydraulic conductivity values were calculated from unadjusted outflow volumes.

Relatively high hydraulic conductivity peaks were observed on the first day of testing. These peaks were discussed in Section 6.2.1. With the exception of plug 1, the hydraulic conductivity curves remain nearly constant after the first few days. Plug 1 exhibits slowly decreasing conductivity values followed by an abrupt increase. The cycle is then repeated. This behavior was attributed to a leaky fitting in the lower end cap. When the water level in the collection buret was at the zero reference height, very little water was lost through the leaky fitting. Water flowing through the plug entered the collection apparatus, thereby increasing the level in the buret. As the level rose, more water was lost through the fitting. When the burets were emptied, higher outflow volumes were measured. The calculated hydraulic conductivities reflect the effect of the leaky fitting. On day 38, tube 1 was isolated from the system and the leaky fitting was replaced. The tube was brought back on-line two days later. The hydraulic conductivity values became constant and similar to those for tubes 2 and 3. Table 6.10(B) presents the hydraulic conductivity values for the system after the leak was repaired.

The hydraulic conductivities shown in Table 6.10 may not reflect the influence of the calcium permeant on the conductivities of the bentonite plugs. The hydraulic conductivity of the plugs, shown in Figures A.16 to A.19, is relatively constant for the full duration of the test. Had the permeant influenced the hydraulic conductivity of the plugs, significant changes in the conductivity should have been observed. It is also possible that no significant conductivity changes will occur in the bentonite plugs as a result of constant pressure

injection testing of the plugs using a 45 ppm calcium permeant. A longer testing period would be required to adequately determine whether a 45 ppm calcium permeant has a significant influence on the hydraulic conductivity of the plugs.

Figures B.28 through B.31 (Appendix B) are mass balance plots for the tests. Relatively high initial influent and effluent peaks similar to those discussed previously are present. The curves for tube 2 converge after approximately 3 days. No convergence is observed in tube 1 until after the leak was repaired. The curves for tube 3 converge very slowly. The mass balance curves for plug 3 suggest that water may have escaped from the collection apparatus. However, no leak was found.

The final moisture contents of the plugs were determined as described in Section 6.2.1 and are presented in Table 6.11. A moisture content variation is observed in each plug, but not the systematic (expected) variation observed in nearly all plugs reported on previously. The moisture content of plug 3 is considerably less than that of plugs 1 or 2, suggesting reduced injection into this plug. This may explain the relatively large differences between the effluent and influent curves for plug 3 compared to those of plugs 1 or 2. The hydraulic conductivities are least for the tube having the smallest moisture content and highest for the tube with the largest moisture content.

6.2.5 Low Sodium Concentration System

Two bentonite plugs were tested using a 39 ppm sodium permeant over a 54-day period under a pressure of approximately 17 kPa (2.5 psi). Plug construction, experimental procedures, and testing apparatus are described in Section 5.2.3. A third plug failed early in the test sequence.

Hydraulic conductivities calculated from measured outflow average approximately 10^{-8} cm/s (Table 6.12). Figures A.20 through A.22 (Appendix A) show the hydraulic conductivity over time. The hydraulic conductivities were calculated from unadjusted outflow volumes.

Relatively high hydraulic conductivity peaks were observed on the first day of testing. These peaks were discussed in Section 6.2.1. This

Table 6.11 Bentonite Plug Moisture Content. Moisture Content at Three Depths of Three Bentonite Plugs After Constant Pressure Injection Testing with 45 ppm Calcium Water

Sample Location	Moisture Content (%) Tube #1	Tube #2	Tube #3
Upper Interface	400	425	294
Midsection	336	387	270
Lower Interface	368	406	282

Table 6.12 Average Hydraulic Conductivity of Bentonite Plugs Using a 39 ppm Sodium Permeant Water

Tube #	Average Hydraulic Conductivity (cm/s)
1[a]	8.1×10^{-9}
2[b]	---
3	1.7×10^{-8}

[a] Small leak observed in collection assembly.

[b] Plug failed upon application of pressure.

experiment suffered numerous problems. Plug 2 failed as soon as pressure was applied to the system, possibly because of an inadequate seal between the plug and the PVC tube wall, or due to a weak spot or fault in the plug, and was taken off-line. A small leak was detected around the swagelok fitting in the lower end cap of tube 1. All attempts to seal the leak failed, resulting in considerably smaller outflow measurements. This tube was kept on-line in spite of the small leak. The hydraulic conductivity values reported for plug 3 were nearly constant after the first four to five days of testing.

Approximately 1.7 and 3.3 pore volumes were collected from plugs 1 and 3, respectively. The volume collected from plug 1 is lower than the actual outflow because of the leak in the lower end cap. The duration of hydraulic conductivity testing may not have been adequate for significant permeant effects to occur. However, significant conductivity changes also may not occur because cation exchange would not be expected to occur with the sodium permeant used in this study.

Figures B.32 through B.34 (Appendix B) are mass balance plots. High peaks are present on the first day. The curves converge after approximately 12 days for tube 3.

The final moisture contents of the plugs were determined as described in Section 6.2.1. Table 6.13 presents the results. Moisture differences are present, but barely so in plug #3, and without a systematic variation in plug #1. The moisture content and the nearly identical influent and effluent volumes for plug 3 suggest that it has attained an equilibrium state.

Table 6.13 Moisture Content of Bentonite Plugs After Constant Pressure Injection Testing with Distilled Water

Sample Location	Moisture Content (%) Tube #1[a]	Tube #2[b]	Tube #3
Upper Interface	318	---	304
Midsection	111	---	293
Lower Interface	215	---	299

[a]Small leak observed in collection assembly.

[b]Plug failed upon application of pressure.

6.3 Consolidation Testing

Section 5.3 describes the consolidation testing objectives, analysis and procedures. Three plugs, constructed from bentonite tablets hydrated with Ogallala groundwater, were consolidated at 34, 69 and 138 kPa (5, 10 and 20 psi). The plugs were consolidated at 34 kPa (5 psi) for 24 days, at 69 kPa (10 psi) for 30 days, and at 138 kPa (20 psi) for 10 days. With the exception of the 138 kPa (20 psi) consolidation period, these periods correspond to the duration of constant injection testing for the Ogallala system. The plugs were consolidated at 138 kPa (20 psi) for only 10 days because of a failure in one or all of the plugs between the tenth and eleventh day of testing. Consolidation was measured as the volume of water drained from the plugs.

Consolidation is usually analyzed by plotting the deformation of the sample against the logarithm of time (Casagrande, 1936). Graphical methods are used to estimate the coefficient of consolidation by approximating the time required for 50 or 90% of total consolidation (Lambe and Whitman, 1969, pp. 408-412; Holtz and Kovacs, 1981, pp. 395-402). These methods utilize the change in slope of the plot as primary consolidation approaches its limit.

The oedometer test described in Section 4.4.2 is the conventional consolidation test. It measures sample deformation using a linear deformation gage. Linear displacement should be proportional to volumetric change, and a similar change in slope should be observed in an outflow volume against the logarithm of time plot. Figures C.1 through C.3 (Appendix C) show the average drained outflow volumes plotted against the logarithm of time for the three plugs at each pressure increment. The curves show no flattening of the slope.

An alternate method of analyzing consolidation is to plot the deformation against the square root of time (Taylor, 1948, pp.

238-242; Holtz and Kovacs, 1981, pp. 400-402). A flattening of the slope should allow approximating the time necessary to achieve a given percentage of consolidation. Figures C.4 through C.15 (Appendix C) show outflow volumes plotted against the square root of time for the three plugs at each pressure increment. No flattening is apparent.

The primary objective of this testing is to estimate what outflow volume can be attributed to consolidation. This volume is then subtracted from the total outflow measured during constant injection testing. The outflow due to consolidation is obtained from outflow volume versus time plots.

Figures C.4 through C.15 show the relationship of volume drained to the square root of time at each test pressure. A significant difference is observed between the plots resulting from 34 kPa (5 psi) and those for 69 and 138 kPa (10 and 20 psi). The consolidation plots at 69 and 138 kPa (10 and 20 psi) are approximately straight lines, with correlation coefficients of 0.96 and 0.99, respectively. The consolidation plot for 138 kPa (20 psi) may have the better apparent correlation because it is based on a substantially smaller test period. Testing at 34 kPa (5 psi) shows a curved plot. This difference may be due to a test interruption from day 7 through day 10, because of an empty nitrogen bottle.

The coefficient of consolidation normally varies with consolidation pressure (Terzaghi, 1943, p. 271; Tavenas et al., 1983). This results from the relationship between pressure and consolidation limits. The consolidation coefficient is calculated for each applied pressure, assuming that consolidation has essentially ended (Holtz and Kovacs, 1981, p. 291). The consolidation plots in Figures C.1 through C.15 (Appendix C) show no indication that consolidation is approaching a limit.

The volume of water drained from the plugs due to consolidation can be determined directly from the consolidation plots. These values, when used in hydraulic conductivity calculations, i.e. subtracted from the total outflow measured in injection tests, gave negative hydraulic conductivity values. A similar observation was made by Kimbrell et al. (1987, p. 252). This suggests that no real flow occurred through the plug. Based on mass balance principles, this is impossible. The values obtained from the consolidation plots were larger than the outflow volumes measured from the corresponding constant injection tests. This suggests rather strongly that this adjustment is not feasible using this type of test. Therefore, all calculated hydraulic conductivity values obtained from constant injection testing used unadjusted outflow volumes in Darcy's law.

Consolidation can also be estimated by comparing the plug heights at the start of a test with those at the end. Table 6.14 summarizes consolidation estimates from measuring plug heights at each pressure increment. Consolidated length is expressed as a percent of the original height. The consolidation shown for the plugs consolidated at 34 kPa (5 psi) may be in error, since the test was interrupted for several days, allowing the plugs to rebound. The plug height estimate

Table 6.14 Bentonite Plug Length Before and After Consolidation Testing

Plug #	Applied Pressure kPa (psi)	Initial Height (cm)	Final Height (cm)	Consolidated Length (Percent of Initial Length)
1	34 (5)	13.4	13.0	97
2	34 (5)	13.5	13.1	97
3	34 (5)	13.2	12.9	98
1	69 (10)	13.0	12.4	95
2	69 (10)	13.0	12.1	93
3	69 (10)	13.1	12.1	92

for consolidation should be considered less accurate than the volumetric estimate, since the plug heights are measured between only two reference points. The volumetric estimate more accurately reflects the volume of the plug being compressed.

The volumetric testing method used in this study is not problem-free. A leak on the pressurized side of the plug drains the nitrogen bottle very rapidly. Lower than planned consolidation pressures may result. A leak on the collection side of the plug may result in low consolidation estimates. Other problems pertaining directly to the use of pressurized nitrogen to consolidate the plugs should also be considered, in particular whether a nitrogen pressurization or injection system can be considered as a consolidation test.

Examination of the plugs during consolidation testing showed that small surface cracks had formed after approximately 15 days of testing at 34 kPa (5 psi). The plug surfaces had also become very dry. The drying may have resulted from plug drainage, suggesting that consolidation drainage occurred from the top of the plug and continued through the plug length. When testing at 34 kPa (5 psi) ended, 80 ml of Ogallala groundwater was added to each plug. Within 24 hours no water was visible on the plug surface and the cracks had closed. Testing at 69 kPa (10 psi) commenced. The surface cracks reappeared within 11 days, and plug drying was observed again. At the end of consolidation at 69 kPa (10 psi), the cracks were visible to a depth of approximately 7.6 cm (3 inches). The original plug heights were approximately 12.7 cm (5 inches). The cracks were approximately 2 to 3 mm (1/10 to 1/8 inch) wide. The cracks closed again when 100 ml of water was added to each tube. Testing at 138 kPa (20 psi) resulted in similar cracks. Figure 6.2 shows the cracks which formed in the plugs during consolidation testing.

The formation of cracks in bentonite plugs during consolidation testing raises doubts about the validity and reliability of the test as

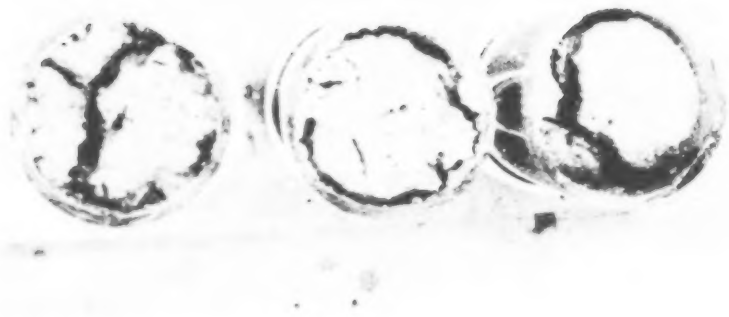

Figure 6.2 Cracks formed in bentonite plugs during "consolidation" testing by applying pressurized nitrogen directly to the plugs.

performed. In the standard oedometer test, a plate covering the entire surface area of the sample is used to apply a uniform load to the sample. Gas pressure applied directly to bentonite plugs may concentrate in the crack and, over time, penetrate the plug. Nonuniform consolidation may result. The experiments clearly reveal the drastic effect of exposing the open end of a bentonite plug to even relatively short-time air pressure.

6.4 Transient Pulse Testing

Transient pulse testing was conducted on the distilled water and Ogallala groundwater plugs previously described under constant pressure injection testing. Transient testing was also conducted on bentonite plugs with a predetermined moisture content. Section 6.4.1 presents the transient test results of the distilled water and Ogallala groundwater systems. Section 6.4.2 presents the test results for bentonite plugs with a predetermined moisture content.

6.4.1 Transient Pulse Testing of Bentonite Plugs with Distilled Water and Ogallala Groundwater Systems

Sections 4.3.2 and 5.3 describe the transient pulse testing objectives, analysis and procedures. Transient testing was conducted prior to any constant pressure injection testing.

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Table 6.15 Bentonite Plug Hydraulic Conductivity Results from Transient Pulse Testing with Distilled Water and with Ogallala Groundwater

Test	Hydraulic Conductivity (cm/s) Plug #1	Plug #2	Plug #3
	Distilled Water:		
1	4.0×10^{-12}	9.5×10^{-12}	1.6×10^{-11}
2	2.2×10^{-11}	2.0×10^{-11}	2.4×10^{-11}
3	1.2×10^{-11}	5.0×10^{-12}	6.0×10^{-11}
	Ogallala Groundwater:		
1	2.3×10^{-11}	6.4×10^{-11}	9.0×10^{-11}
2	2.5×10^{-12}	8.6×10^{-12}	1.4×10^{-11}
3	6.8×10^{-12}	9.1×10^{12}	8.7×10^{-12}

Hydraulic conductivities determined from transient testing range from 10^{-11} to 10^{-12} cm/s for the distilled water and Ogallala groundwater plugs. Figures D.1 through D.8 (Appendix D) show the decay of hydraulic head in the upstream reservoir over time. These experimental curves closely approximate the theoretical type curve obtained from the early time approximation (Figure D.9). Table 6.15 summarizes the results. The average conductivity for the distilled water system is 1.9×10^{-11} cm/s and 2.5×10^{-11} cm/s for the Ogallala groundwater system. The conductivity values for both systems are essentially the same.

The hydraulic conductivity values for transient pulse testing are several orders of magnitude smaller than the values obtained from constant injection testing of the same plugs. These transient values are at the lower end of conductivity values reported in the literature. The disparity between the hydraulic conductivity values obtained from constant injection and transient testing methods might not be as great if the outflow volumes were adjusted for consolidation, which is likely to be most significant in the constant injection test.

A second hydraulic property, the specific storage (Sections 4.3 and 5.3), was determined for the bentonite plugs. The Hsieh et al. (1981)

Table 6.16 Bentonite Plug Specific Storage and Compressive Storage Determined from Transient Pulse Testing with Distilled Water and with Ogallala Groundwater

System	Specific Storage (cm^{-1})	Compressive Storage, Upper Reservoir (cm^2)
Distilled Water	7.1×10^{-3}	9.2×10^{-3}
Ogallala Groundwater	3.4×10^{-2}	3.7×10^{-2}

early time solution does not give an independent value for specific storage. The product of hydraulic conductivity and specific storage is calculated from the type curve matching method outlined by Neuzil et al. (1981). If a value for β is known, Equation (4.12) can be rearranged, allowing calculation of the specific storage of the sample. The hydraulic conductivity of the sample can then be calculated by substituting the specific storage value into Equation (4.11). The compressive storage of the upper reservoir for both systems was determined experimentally. The β value was determined from the type curve that gave the best fit with the decay curves. A value of 1000 was used for β. Table 6.16 presents the specific storage and compressive storage results for both the distilled water and Ogallala groundwater systems.

The specific storage determined from transient pulse testing for the Ogallala groundwater system was compared with a value calculated from consolidation data and the equation which defines specific storage. The specific storage S_s of a material is given by (Freeze and Cherry, 1979, p. 59):

$$S_s = \rho g(\alpha + n\beta) \qquad (6.2)$$

where n = porosity
α = compressibility of the medium,
β = fluid compressibility, and
ρ = density.

The compressibility of the medium is given by (Freeze and Cherry, 1979, p. 54):

$$\alpha = (-dV/V)/(d\sigma_e) \qquad (6.3)$$

where V = the volume of the medium, and
$d\sigma_e$ = the change in effective stress.

When the total stress does not change with time, the effective stress at any point in the system, and the resulting volumetric deformations are controlled by the fluid pressure at that point (Freeze and Cherry, 1979, p. 53). Changes in the effective stress at a point are given by:

$$d\sigma_e = -\rho g dh \qquad (6.4)$$

where h is the pressure head, considered a constant at a point. Equation (6.3) can be expressed as:

$$\alpha = (db/b)/(\rho g dh) \qquad (6.5)$$

where b is the original plug height (Freeze and Cherry, 1979, p. 54).

If the deformation of the sample determined from the independent consolidation test, described in Sections 5.3 and 6.3, is used in Equation (6.5), the compressibility of the medium is estimated as 2×10^{-6} m^2/N (in the upper range of values listed for clay by Freeze and Cherry, 1979, p. 55). The specific storage is then calculated using equation (6.3) and 4.4×10^{-10} m^2/N for the compressibility of water. The porosity of the clay is estimated at 40 percent. Davis (1969) gives porosities for clays ranging from 40 to 70 percent. The specific storage thus calculated is 2×10^{-4} cm^{-1}.

A major assumption made in determining the compressibility of the medium is that hydraulic and mechanical (deformational) equilibrium has been achieved. Hydraulic equilibrium is achieved when the change in the pore pressure within the sample is zero. This condition was not attained for the consolidation testing. If the consolidation testing had been continued until hydraulic equilibrium was established, the calculated specific storage of the bentonite may have approached the value obtained from the Hsieh et al. (1981) solution.

Air bubbles trapped in voids within the clay plug also affect the results of transient pulse testing. Transient pulse testing requires that a pressure slug or pulse be introduced to a reservoir on one side of the material tested. The decay of the pressure pulse is monitored and the hydraulic properties of the material are determined through curve matching. Air-filled voids retard fluid flow in a material. Upon application of the pressure pulse, the air bubbles can be expected to decrease in volume, thereby allowing for a greater fluid flow. As the pressure pulse decays, air bubbles expand, resulting in restricted fluid flow. It is unclear what the overall effects air bubbles trapped within the test medium have on the decay curves and the resultant hydraulic conductivities.

Table 6.17 Hydraulic Conductivity Values Obtained From Transient Pulse Testing with Distilled Water and Ogallala Groundwater of Bentonite Plugs with Predetermined Initial Moisture Contents

Initial Moisture Content	Distilled Water System			Ogallala Groundwater System		
	Hydraulic Conductivity	Specific Storage		Hydraulic Conductivity	Specific Storage	
(%)	(cm/s)	(cm^{-1})	β	(cm/s)	(cm^{-1})	β
75	8.9×10^{-8}	3.0×10^{-3}	1	5.5×10^{-8}	1.9×10^{-3}	1
100	Pressure pulse punctured plug.			Test failed.		
125	5.9×10^{-9}	6.1×10^{-3}	5	2.4×10^{-8}	2.7×10^{-4}	0.1

6.4.2 Transient Pulse Testing of Bentonite Plugs with Predetermined Moisture Contents

Sections 4.3.2 and 5.3 describe the transient pulse test objectives, analysis and procedures.

Hydraulic conductivities determined from transient testing ranged from 10^{-8} to 10^{-9} cm/s for the distilled water and Ogallala groundwater plugs. Figures D.10 through D.14 (Appendix D) show the decay of hydraulic head in the small upstream reservoir over time, while Figures D.15 through D.19 show hydraulic head decay in the large upstream reservoir. Pressure buildup in the downstream reservoirs is also shown. Each experimental data curve closely approximates one of the theoretical type curves shown in Figures D.25 through D.27. Table 6.17 summarizes the results of transient pulse testing. Specific storage and β values for each plug are included. The β values were determined from curve matching techniques presented by Neuzil et al. (1981).

Hydraulic conductivities were determined from experimental decay curves in the small upstream reservoirs only. The small downstream reservoirs were insufficient for adequate decay to occur when the pressure pulse was introduced into the large upstream reservoirs. Figures D.15 through D.19 (Appendix D) illustrate the hydraulic head decay in the large upstream reservoirs. Ideally, fifty percent hydraulic head decay in the upstream reservoir is required before the hydraulic conductivity of a material can be determined. The largest decay into a small downstream reservoir, approximately 32 percent, occurred during testing of a distilled water plug having a moisture content of 75 percent. Head decays of approximately 4 to 15 percent occurred in the remaining plugs.

Table 6.18 Specific Storage Values Obtained From Transient Pulse Testing with Distilled Water and Ogallala Groundwater of Plugs with Predetermined Initial Moisture Contents

Initial Moisture Content (%)	Specific Storage (cm^{-1}) Distilled Water System		Ogallala Groundwater System	
	Two-Test Method	Curve-Matching Method	Two-Test Method	Curve-Matching Method
75	3.7×10^{-3}	3.0×10^{-3}	2.3×10^{-3}	1.9×10^{-3}
100	--	--	--	--
125	1.1×10^{-3}	6.1×10^{-3}	1.5×10^{-3}	2.7×10^{-4}

The experimental decay curves obtained from this phase of transient pulse testing showed an unexpected pressure buildup in the upstream reservoirs near the end of each testing episode. Figures D.10 through D.14 show a gradual hydraulic head decay in the upstream reservoir accompanied by a smaller hydraulic head buildup in the downstream reservoir. The large ratio between the downstream and upstream reservoir volumes would account for the small increase of hydraulic head in the downstream reservoir. After approximately 1100 minutes, a rapid buildup in hydraulic head is observed in the upstream and downstream reservoirs. Storage characteristics of bentonite may, in part, explain this behavior.

The hydraulic head in the small upstream reservoirs experienced approximately 30 percent decay after 100 minutes while the buildup of head in the downstream reservoirs was less than 5 percent. This suggests, assuming no pressure loss had occurred due to leaks, that the pore pressure within the bentonite plug has increased. The pore pressure continues to increase until it exceeds that of the fluid reservoirs. A gradient reversal occurs followed by a rapid increase in hydraulic head in both the upstream and downstream reservoirs.

Hydraulic conductivities determined from transient testing of bentonite plugs with a predetermined moisture content were several orders of magnitude higher than those obtained from transient testing of plugs used in constant pressure injection testing. Transient testing of the former resulted in conductivities ranging from 10^{-8} to 10^{-9} cm/s, while the latter yielded conductivities ranging from 10^{-11} to 10^{-12} cm/s. Differences in plug construction may account for the wide range of conductivities between each set of plugs. Bentonite plugs utilized in constant pressure injection testing were constructed "in situ" by dropping American Colloid Volclay tablets through a column of water and allowing the tablets to hydrate and swell (Section 5.2.1). The plugs with specific moisture contents were constructed by placing sufficient

volumes of premixed clay in a mold (Section 5.3). It is likely that plugs constructed using this technique had a larger pore volume than those constructed in situ.

Transient pulse testing of bentonite plugs constructed from premixed clay with a moisture content of 100 percent failed. The plug contructed from clay hydrated with distilled water was immediately punctured upon introduction of the pressure pulse. The plug constructed from clay hydrated with Ogallala groundwater allowed 23 percent of the pressure pulse to decay from the upstream reservoir to the downstream reservoir in the first 20 minutes of the test. Only 14 percent decay occurred during the 1100 minutes following the initial pressure decrease. The experiment was repeated using a different set of bentonite plugs. The second experiment yielded similar results.

A secondary objective of transient pulse testing was to determine specific storage values of bentonite plugs. The two-test procedure presented by Neuzil et al. (1981) was conducted to determine the specific storage of bentonite plugs having perdetermined moisture contents. Table 6.18 summarizes the specific storage results. Also shown are the specific storage values obtained from curve matching. These compare favorably with a theoretical value calculated from consolidation data (Section 6.4.1).

Transient pulse testing may develop into a quick and reliable method for determining the hydraulic properties of low permeability clays such as bentonite. However, a far more exhaustive testing program will be required to demonstrate its validity. For in-situ testing of actual plugs in particular, the need for saturation may induce complications or preliminary long-term injection, or testing to assure steady-state conditions before initiating intentional transient (pulse) testing. Any of these approaches might somewhat defeat the prime advantage of "rapid" testing of a low-permeability medium.

CHAPTER SEVEN

SUMMARY, CONCLUSIONS AND RECOMMENDATIONS

7.1 Summary

Laboratory testing was conducted to determine the effect of permeant chemistry on the hydraulic conductivity of bentonite. The hydraulic conductivity was determined using two methods. Constant pressure injection testing was performed with five different permeants, transient pulse testing with two different waters.

The following discussions, experiments and analyses are included:

1) The structure and characteristics of montmorillonite are summarized. Structural considerations include the configuration and bonding of the unit layers constituting the montmorillonite mineral. Cation exchange, isomorphic substitution, diffuse double layer, and swelling characteristics are discussed.

2) Properties of the bentonite used in this study are presented.

3) Physical and chemical properties of permeants which affect the hydraulic conductivity of bentonite are reviewed.

4) Characteristics of the water permeants are presented.

5) The hydraulic conductivity of three 10-cm (4-inch) diameter bentonite plugs has been determined using constant pressure injection of a distilled water permeant.

6) The hydraulic conductivity of three 10-cm (4-inch) diameter bentonite plugs has been determined using constant pressure injection of a groundwater (Ogallala) permeant.

7) Three 10-cm (4-inch) diameter bentonite plugs have been "consolidated" under pressurized nitrogen.

8) Swelling characteristics of bentonite mixed with distilled water and with a groundwater are illustrated.

9) Constant pressure injection testing is conducted with a high concentration (1000 ppm) calcium solution on bentonite plugs constructed at three different initial moisture contents

10) The hydraulic conductivity of three 5-cm (2-inch) diameter bentonite plugs has been determined using constant pressure injection testing with a low concentration (45 ppm) calcium solution.

11) The hydraulic conductivity of three 5-cm (2-inch) diameter bentonite plugs has been determined using constant pressure injection with a low concentration (39 ppm) sodium solution.

12) Transient pulse testing on the 10-cm (4-inch) diameter bentonite plugs used in constant pressure injection testing for both the distilled water and groundwater systems has been used to determine the hydraulic conductivity and specific storage of the clay.

7.2 Conclusions

This discussion is specific to the tested American Colloid Volclay bentonite tablets. Commercial bentonites include a wide variety of chemical and mineral compositions (e.g. Sawyer and Daemen, Table 4.1). It may not be adequate to rely on generic bentonite studies when evaluating the suitability of a bentonite for a particular application.

Bentonite tablets provide a convenient means for forming a low-permeability barrier. They have aspects which cause difficulties in performance testing. Constructing a plug by dropping bentonite tablets through water does not allow control of plug density or homogeneity. Visual inspection of plugs constructed in this manner reveals nonuniform swelling and pockets or voids within the plug. These may provide a preferential flowpath. Incomplete tablet hydration and swelling may also result from this installation method. Prior to installation, plug volumes can only be estimated. Final volumes depend on the availability of water to the tablets as well as on the chemical composition of the water. The tablets may retain some air-filled void space, even when hydrated. They are, therefore, not completely saturated. The moisture content changes with experimental conditions. Mass balance plots suggest that a flow equilibrium condition is achieved between the bentonite plug and the injection pressure after relatively short testing periods. Constant pressure injection testing of bentonite initiates consolidation drainage of the plugs. Adjustment of outflow volumes used to calculate hydraulic conductivity may be necessary to account for this consolidation drainage, particularly when long-term constant pressure injection test methods are employed.

Constant pressure injection testing of bentonite plugs constructed from American Colloid Volclay tablets hydrated with distilled water and using a distilled water permeant resulted in average hydraulic conductivities of 1.9×10^{-8} cm/s at 34 kPa (5 psi), 1.5×10^{-8} cm/s at 69 kPa (10 psi), and 1.0×10^{-8} cm/s at 138 kPa (20 psi) injection pressure. The small decrease of hydraulic conductivity with increasing pressure suggests that consolidation may influence the hydraulic conductivity.

Similar testing of plugs constructed, hydrated, and tested with groundwater from the Ogallala aquifer in northern Texas resulted in average hydraulic conductivities of 8.5×10^{-9} cm/s at 34 kPa (5 psi), 8.4×10^{-9} cm/s at 69 kPa (10 psi), and 5.8×10^{-9} cm/s at 138 kPa (20

psi). A decrease in hydraulic conductivity with increasing pressure was observed. However, testing at 34 and 69 kPa (5 and 10 psi) resulted in nearly identical hydraulic conductivity values. Several mechanisms may be responsible for the observed conductivity behavior of the bentonite plugs hydrated and tested with Ogallala groundwater. Frenkel et al. (1978) suggest that plugging of pores by dispersed clay particles is a major cause of reduced soil hydraulic conductivity. Rowell et al. (1969) showed that the solution concentration at which clay disperses depends on the applied mechanical stress. Application of sufficient mechanical stress to a clay can cause dispersion at solution concentrations not otherwise associated with dispersion.

A third mechanism may help to explain the differences in hydraulic conductivity behavior between the distilled water and Ogallala groundwater. The reduced swelling of the Ogallala groundwater system, when compared to the distilled water system, resulted in smaller plug volumes and, consequently, reduced pore volumes. These plugs were not as sensitive to lower injection pressures and did not experience significant consolidation until 138 kPa (20 psi) was applied.

Finally, a high particulate concentration in the groundwater may account for these unexpected results. During plug construction, colloidal matter in the groundwater may have lodged in flow channels, thereby restricting fluid flow.

Constant pressure injection testing of three distilled water plugs using a permeant with 0 SAR and a total salt concentration of 50 meq/l (1000 ppm calcium) resulted in hydraulic conductivities of 6.4×10^{-9} cm/s for a plug with an initial moisture content of 75%, 9.2×10^{-9} cm/s for a plug with an initial moisture content of 100%, and 1.3×10^{-8} cm/s for a plug with an initial moisture content of 125%. Initial moisture content may affect the hydraulic conductivity of the plugs for the given set of experimental conditions. Throughout the 44-day test, a relation between initial moisture content and hydraulic conductivity was observed for each plug. However, the primary objective, cation exchange, may not have been achieved. Had sodium ions adsorbed on the clay mineral surfaces been replaced by calcium, the hydraulic conductivity should have increased. The volume of permeant injected into each plug was insufficient to displace all the distilled water originally occupying the pore space within the plugs and collection reservoirs. A chemical analysis performed on the influent and effluent near the end of the test period showed similar calcium concentrations in each fluid, indicating no cation exchange, or that channeling may have occurred. The small injection volumes and the relatively constant hydraulic conductivity observed in each plug do not support the latter. Contamination of the sample during the chemical analysis is a possible explanation.

Constant pressure injection testing of three distilled water plugs using a permeant with 0 SAR and a total salt concentration of 2.2 meq/l (45 ppm calcium) resulted in an average hydraulic conductivity of 1.7 x

10^{-8} cm/s. Similar testing of a set of three plugs using a permeant having a SAR of ∞ and total salt concentration of 1.7 meq/ℓ (39 ppm sodium) also resulted in a hydraulic conductivity of 1.7×10^{-8} cm/s. Because of a plug failure and because of excessive leakage in an additional tube, the hydraulic conductivity reported for the 1.7 meq/l (low sodium) system reflects only one tube. It is likely that the volume of permeant injected during the 54 day test period was insufficient to displace all the distilled water originally occupying the pore space of the plugs and collection reservoirs. Anderson and Jones (1983) suggest that it may be necessary to pass more than one or even two pore volumes of leachate through a barrier material to assess the effects of the leachate on the barrier material.

The results of bentonite plug constant pressure injection testing hint at the complexity of the material. Many factors influence its behavior, as well as a variety of environmental conditions. Factors influencing the hydraulic conductivity of bentonite include consolidation and swelling, degree of saturation, particle dispersion, blockage or restriction of pore volumes, and clay composition content (Lambe and Whitman, 1969, pp. 283-292; Rowell et al., 1969; Frenkel et al., 1978). There is less agreement on the relative importance of each factor for a given set of conditions. Generally, hydraulic conductivity decreases for high SAR values and decreasing salt concentrations, due to enhanced swelling and dispersion (e.g. Figure 7.1). The relative influence of swelling and dispersion on hydraulic conductivity with regard to solution concentration and composition is not clearly understood. ESP also affects the hydraulic conductivity of a soil. The conductivity decreases as ESP increases, provided the permeant concentration remains below a critical threshold level (Quirk and Schofield, 1955). This threshold level must be determined for each soil. Particle accumulation within soil pores may cause reductions in hydraulic conductivity. Solution chemistry may influence the physical and chemical forces which control colloid migration and attachment to porous media.

The hydraulic conductivities of the bentonite plugs used in constant pressure injection testing of the distilled water and Ogallala groundwater systems were also determined using a transient pulse test method originally designed for low permeability rocks. This transient pulse testing yielded hydraulic conductivities ranging from 10^{-11} to 10^{-12} cm/s. These values are orders of magnitude less than those obtained from constant pressure injection testing of the same plugs. The differences may be reduced if outflow volumes are adjusted for consolidation. Since some doubt exists as to the degree of saturation of the tested plugs, hydraulic conductivities obtained from constant pressure injection testing may be high. Using oedometer testing (Holtz and Kovacs, 1981, pp. 402-403) to determine the hydraulic conductivity of bentonite would account for consolidation drainage and may yield more representative conductivity values.

Transient pulse testing was also performed on bentonite plugs with specific inital moisture contents. Hydraulic conductivities calculated

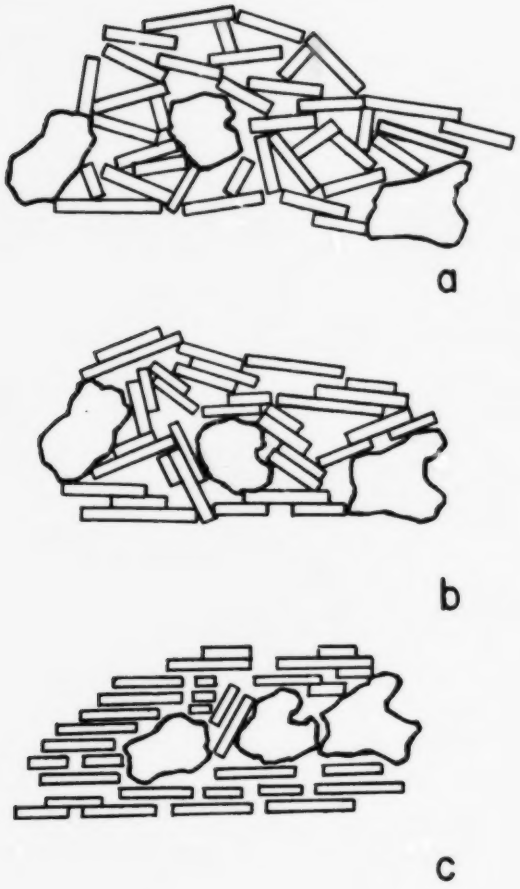

Figure 7.1 Clay cardhouse structure as influenced by water composition and compaction.

a. Undisturbed - Salt Water Deposit
b. Undisturbed - Fresh Water Deposit
c. Remolded

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from this testing ranged from 10^{-8} to 10^{-9} cm/s. These values are orders of magnitude higher than those obtained from transient testing of plugs used in constant pressure injection testing. Differences in plug construction may account for the large difference of conductivities between each set of plugs.

Transient pulse testing was also used to determine specific storage of values for the bentonite plugs. Specific storage calculated from transient testing is of the order of 10^{-3} cm^{-1}, about an order of magnitude larger than a value calculated theoretically from estimated pore space and bentonite compressibilty.

7.3 Further Research Recommendations

Areas requiring additional research include: 1) implications of the construction of inhomogeneous bentonite plugs, 2) migration of colloidal matter and particles through porous media, and 3) development of relatively quick, reliable methods to determine hydraulic properties of low permeability clays.

1) Dropping bentonite tablets through water, a standard well sealing procedure, produces a non-homogeneous plug. Although the resulting plugs tend to have a very low hydraulic conductivity, they also tend to contain voids, weak spots, and zones of incomplete tablet hydration and swelling. These factors may affect the hydraulic conductivity of the plug, and presumably also its strength. They make representative testing difficult, and certainly introduce scaling sensitivity, making application of results from tests on frequently used extremely small and uniform bentonite samples to actual plugs constructed by this method rather questionable. The ability to test bentonite with confidence may be instrumental in evaluating bentonite for specific applications such as borehole sealing, and for example is explicitly required in the NRC Rule 10CFR60 (U.S. Nuclear Regulatory Commission, 1983a, §142) and GTP on borehole and shaft sealing (U.S. Nuclear Regulatory Commission, 1986).

2) Theories for predicting particle and colloid migration through porous media have been developed in the filtration literature. Solids much smaller than pore sizes can migrate into and lodge within the pores and reduce the hydraulic conductivity. Physical and chemical forces, as well as physical straining of the particles by the porous medium, control migration. Because many groundwaters may contain solids, it would be desirable to investigate systematically the influence thereof on seal conductivity. This could be initiated by an evaluation of the applicability of the more fundamental understanding that has been developed in the filtration literature of the physical and chemical forces that affect particle migration through and attachment to clay, specifically bentonite. This could be accompanied by a systematic experimental study of the influence of solids carried by the water on flow through bentonitic seals. Such an approach would include an investigation of likely particle contents in repository location groundwaters. On the basis of such a determination, one could proceed by studying the effect of particle content, size distribution,

and possibly shape. As always when dealing with bentonitic seals, it would be necessary to select controlled representative emplacement procedures as well as water chemistry.

3) Testing methods for determining the hydraulic parameters of low permeability clays need improvement, or, at the very least, much more convincing validity demonstrations than presently available. A variety of methods are available for testing soils and low permeability rocks. However, there is little agreement as to the best method of testing low permeability clays. Standard soil testing methods such as falling head and constant head tests may not account for significant processes affecting the hydraulic conductivity of bentonite. Additionally, they assume saturation and require extensive test periods. A faster yet reliable method for testing low permeability clays is needed. Testing bentonite using transient pulse test procedures might be developed into an effective method for determining the hydraulic properties of low permeability clays. Much more research, including development of theoretical analysis procedures as well as experimental methods, is required before a reliable assessment of the validity of the method can be made. This is likely to deserve considerable priority given the NRC requirement for seal performance demonstration testing (e.g. 10 CFR 60 §142; U.S. Nuclear Regulatory Commission, 1987; GTP on Borehole and Shaft Sealing, Sections 4.2, 4.5, 6.0, U.S. Nuclear Regulatory Commission, 1986).

4) The consequences of emplacing bentonite (or bentonite/crushed rock) seals or backfill in an unsaturated environment, or the effects of drying induced by emplacement relatively close to waste, i.e. sufficiently close for the bentonite to be affected by the drying induced by the thermal pulse, need to be investigated. Even relatively short-term subjecting of bentonite to air or nitrogen pressure results in rapid drying, and the development of shrinkage cracks. Although during slow resaturation a reconstitution of the low permeability predyring bentonite plug is likely, it is far from obvious what the response might be to a relatively rapid inflow of water to such a system, given that the geometry would seem to be an open invitation to piping, channeling, particle transport, and similar phenomena.

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Znidarcic, D., 1982, "Laboratory Determination of Consolidation Properties of Cohesive Soil," Ph.D. Dissertation, University of Colorado, Boulder, 172 pp.

APPENDIX A

HYDRAULIC CONDUCTIVITY PLOTS OBTAINED FROM CONSTANT PRESSURE INJECTION TESTING OF BENTONITE PLUGS

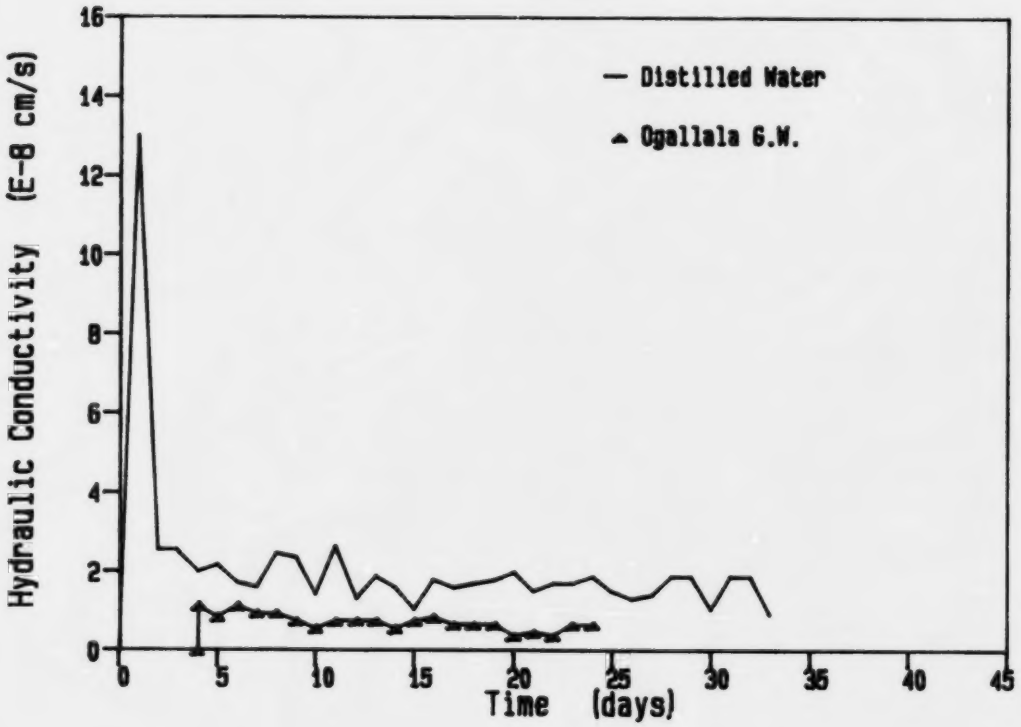

Figure A.1 Hydraulic conductivity of plug 1 determined by constant pressure injection with distilled water and with Ogallala groundwater at 34 kPa (5 psi). Hydraulic conductivity multiplier = 10^{-8} cm/s.

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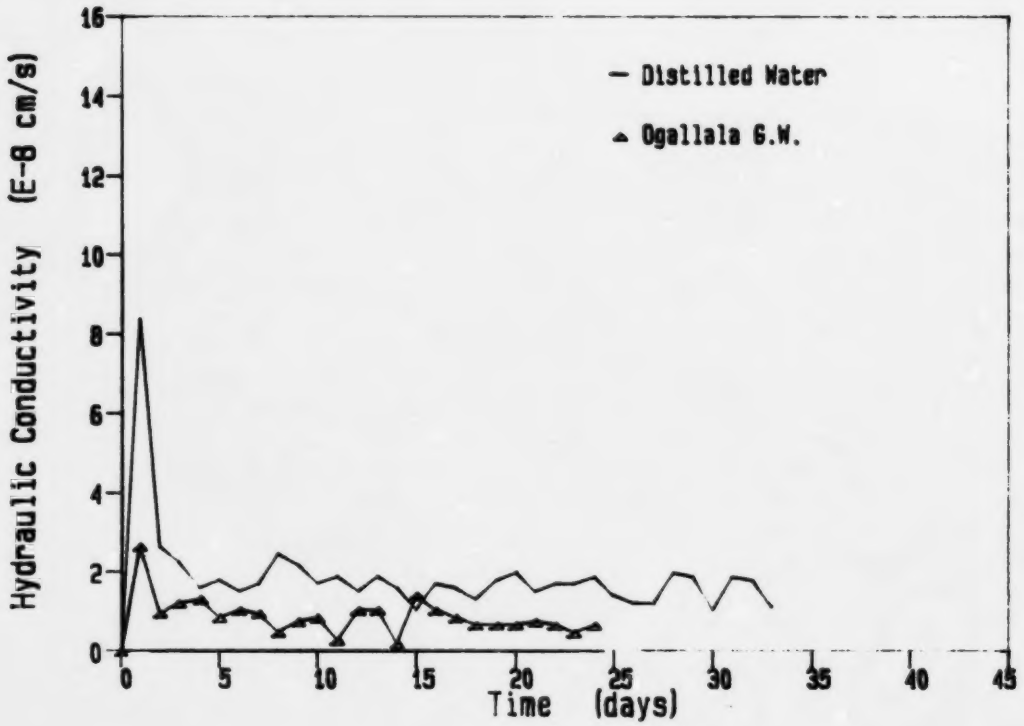

Figure A.2 Hydraulic conductivity of plug 2 determined by constant pressure injection with distilled water and with Ogallala groundwater at 34 kPa (5 psi). Hydraulic conductivity multiplier = 10^{-8} cm/s.

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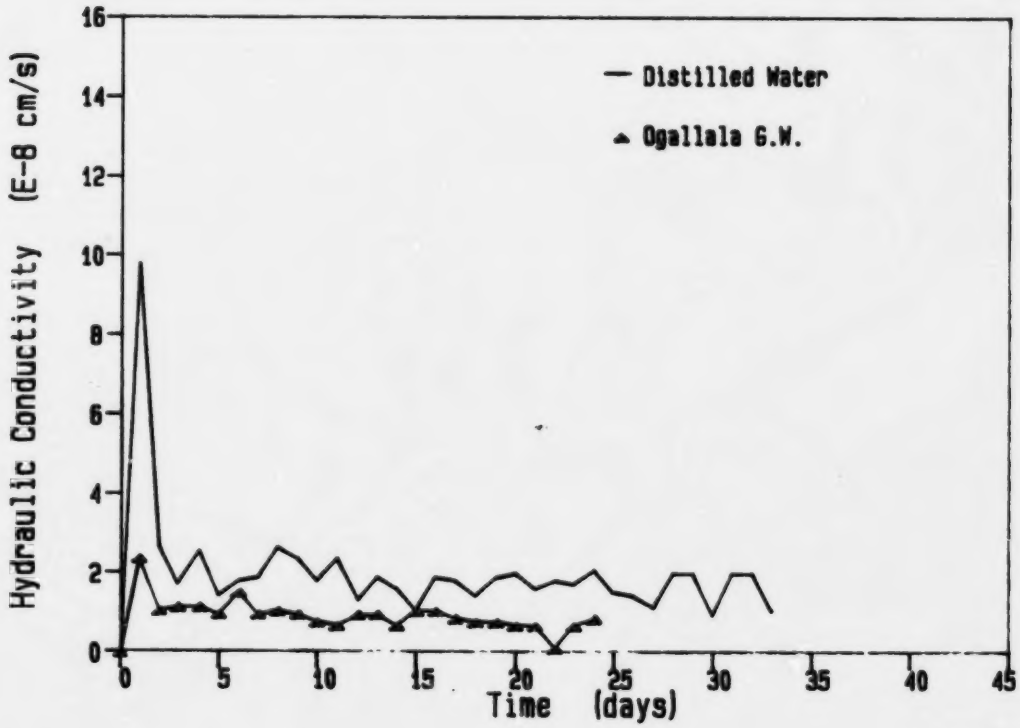

Figure A.3 Hydraulic conductivity of plug 3 determined by constant pressure injection with distilled water and with Ogallala groundwater at 34 kPa (5 psi). Hydraulic conductivity multiplier = 10^{-8} cm/s.

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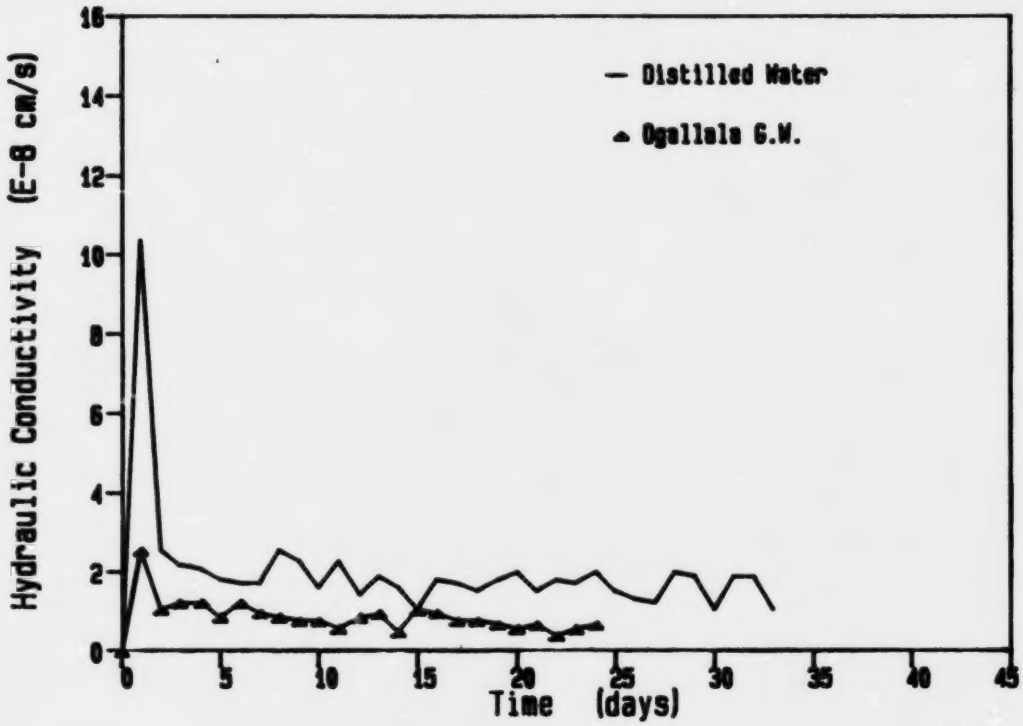

Figure A.4 Average hydraulic conductivity of 3 plugs, determined by constant pressure injection with distilled water and with Ogallala groundwater at 34 kPa (5 psi). Hydraulic conductivity multiplier = 10^{-8} cm/s.

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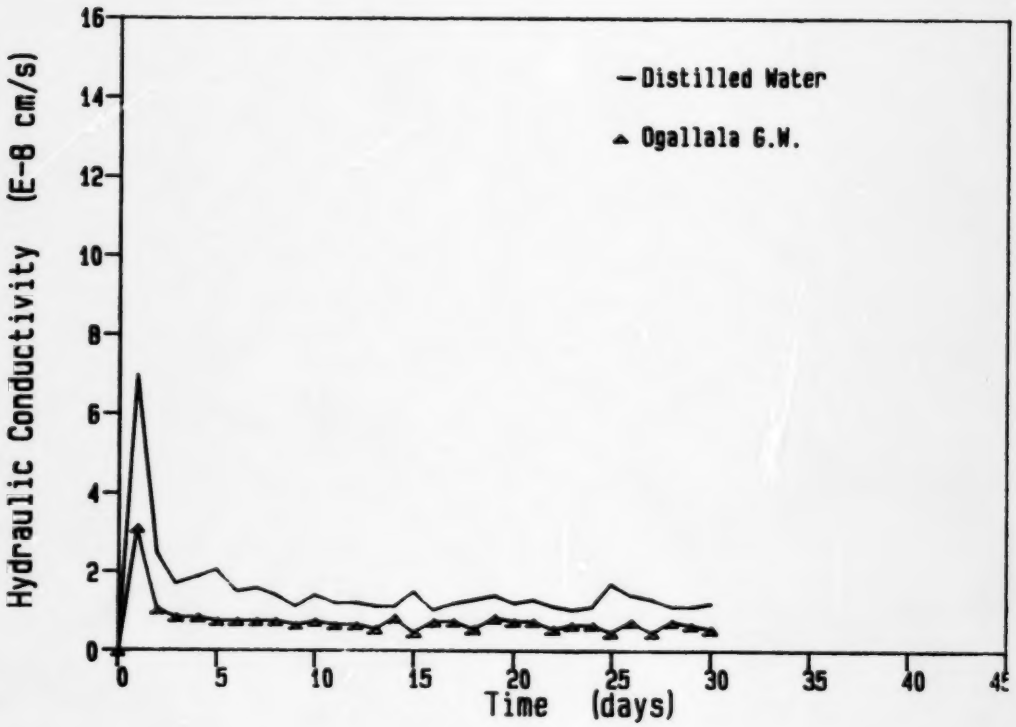

Figure A.5 Hydraulic conductivity of plug 1 determined by constant pressure injection with distilled water and with Ogallala groundwater at 69 kPa (10 psi). Hydraulic conductivity multiplier = 10^{-8} cm/s.

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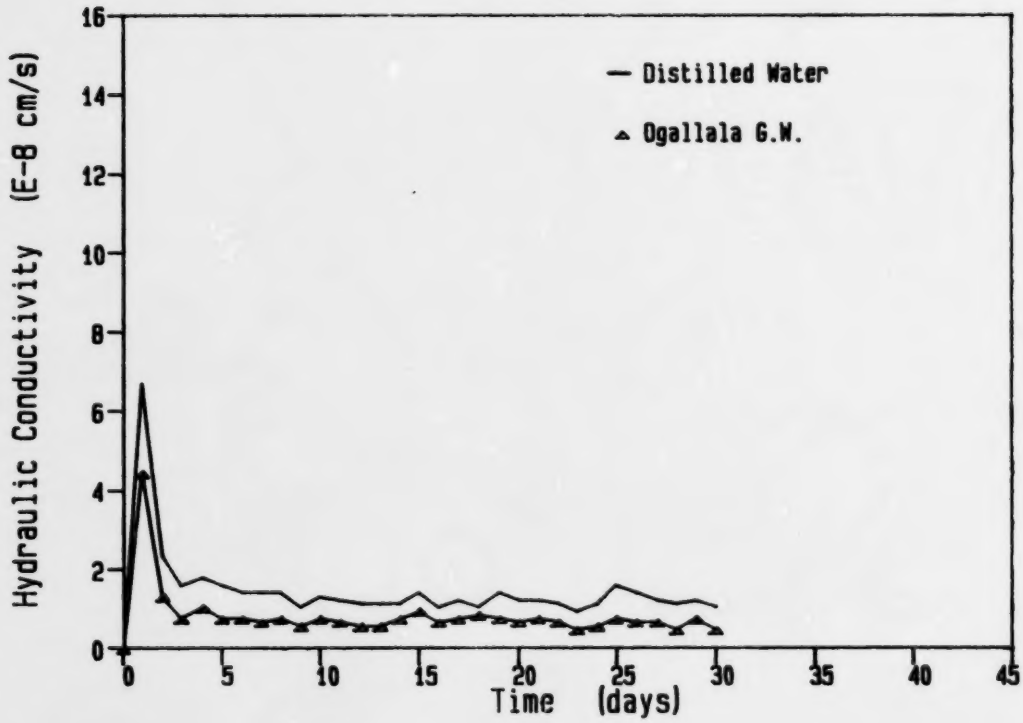

Figure A.6 Hydraulic conductivity of plug 2 determined by constant pressure injection with distilled water and with Ogallala groundwater at 69 kPa (10 psi).

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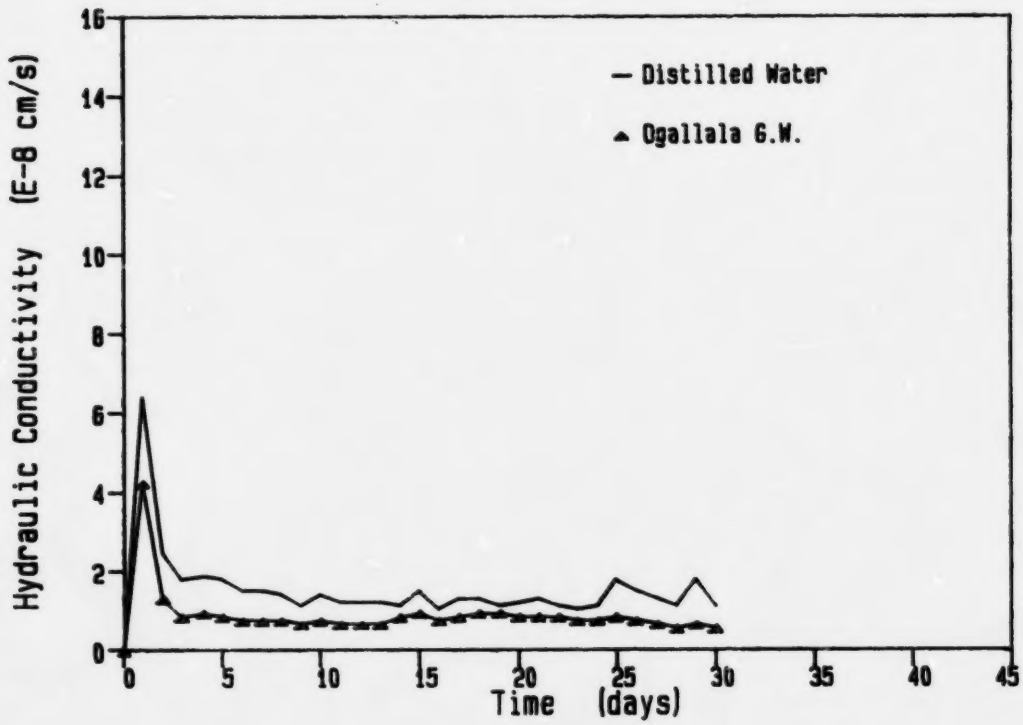

Figure A.7 Hydraulic conductivity of plug 3 determined by constant pressure injection with distilled water and with Ogallala groundwater at 69 kPa (10 psi). Hydraulic conductivity multiplier = 10^{-8} cm/s.

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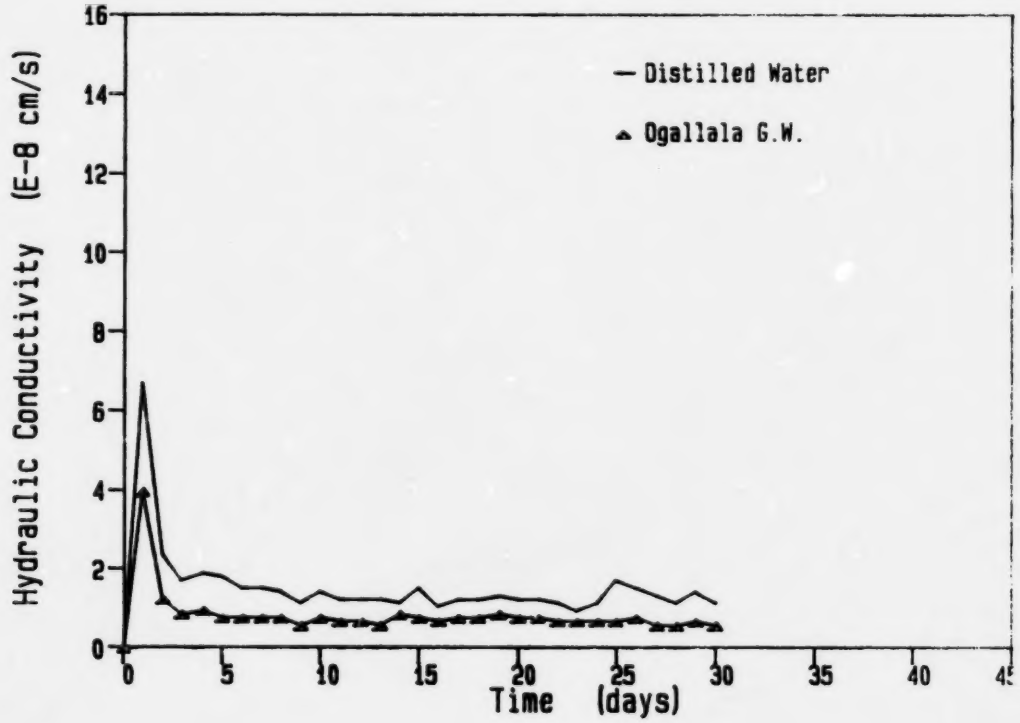

Figure A.8 **Average hydraulic conductivity of 3 bentonite plugs determined by constant pressure injection with distilled water and with Ogallala groundwater at 69 kPa (10 psi). Hydraulic conductivity multiplier = 10^{-8} cm/s.**

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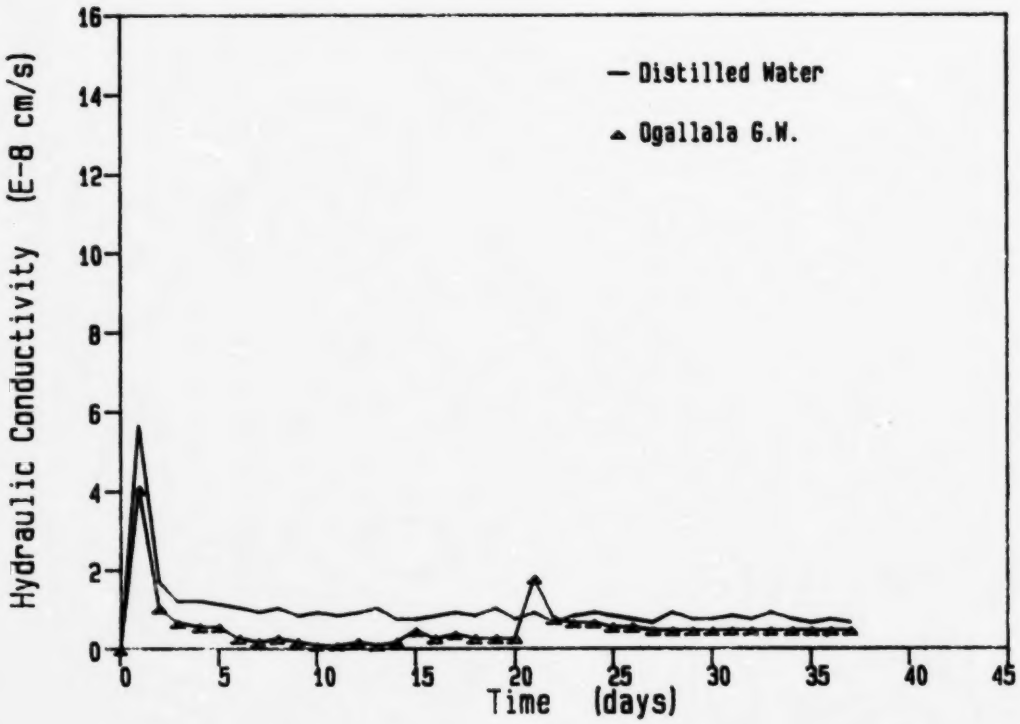

Figure A.9 Hydraulic conductivity of plug 1 determined by constant pressure injection with distilled water and with Ogallala groundwater at 138 kPa (20 psi). Hydraulic conductivity multiplier = 10^{-8} cm/s.

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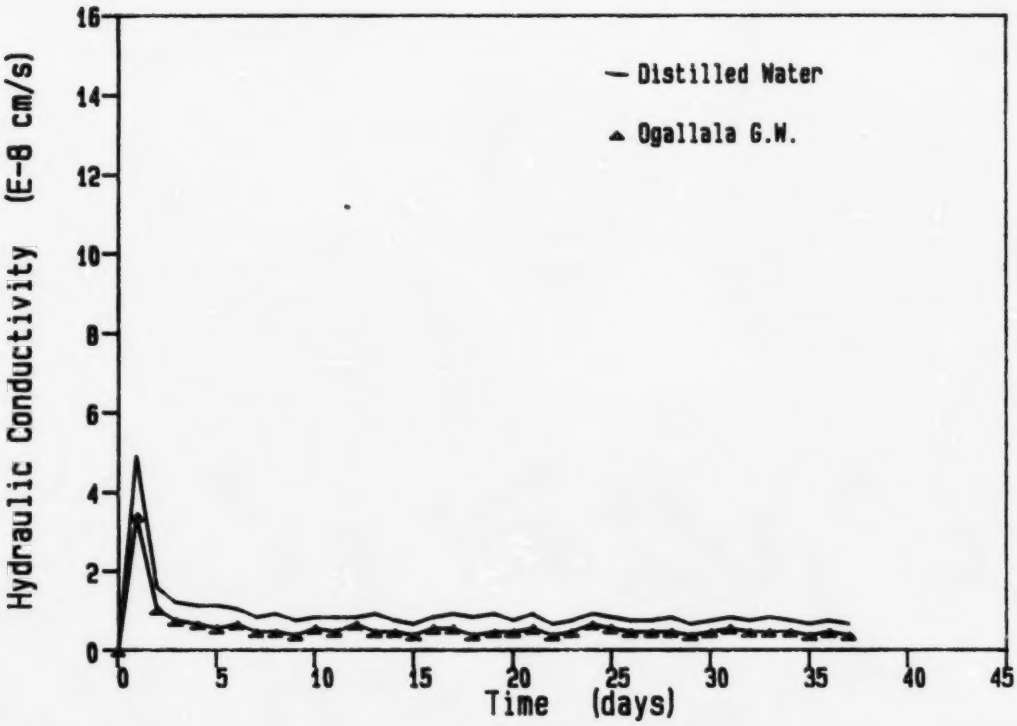

Figure A.10 Hydraulic conductivity of plug 2 determined by constant pressure injection with distilled water and with Ogallala groundwater at 138 kPa (20 psi). Hydraulic conductivity multiplier = 10^{-8} cm/s.

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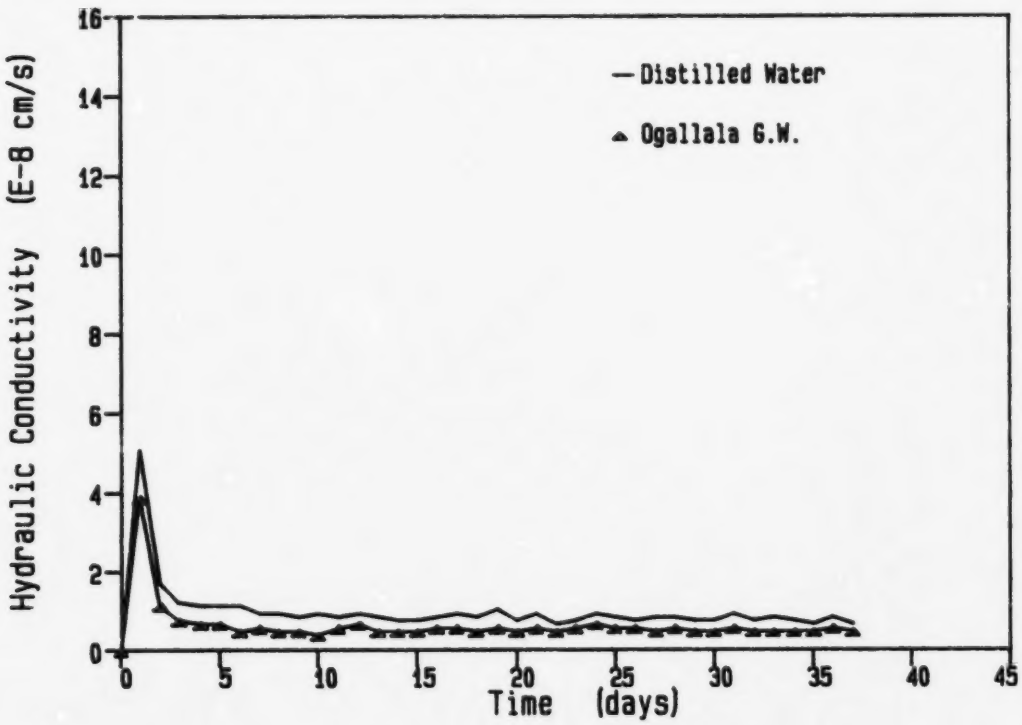

Figure A.11 Hydraulic conductivity of plug 3 determined by constant pressure injection with distilled water and with Ogallala groundwater at 138 kPa (20 psi). Hydraulic conductivity multiplier = 10^{-8} cm/s.

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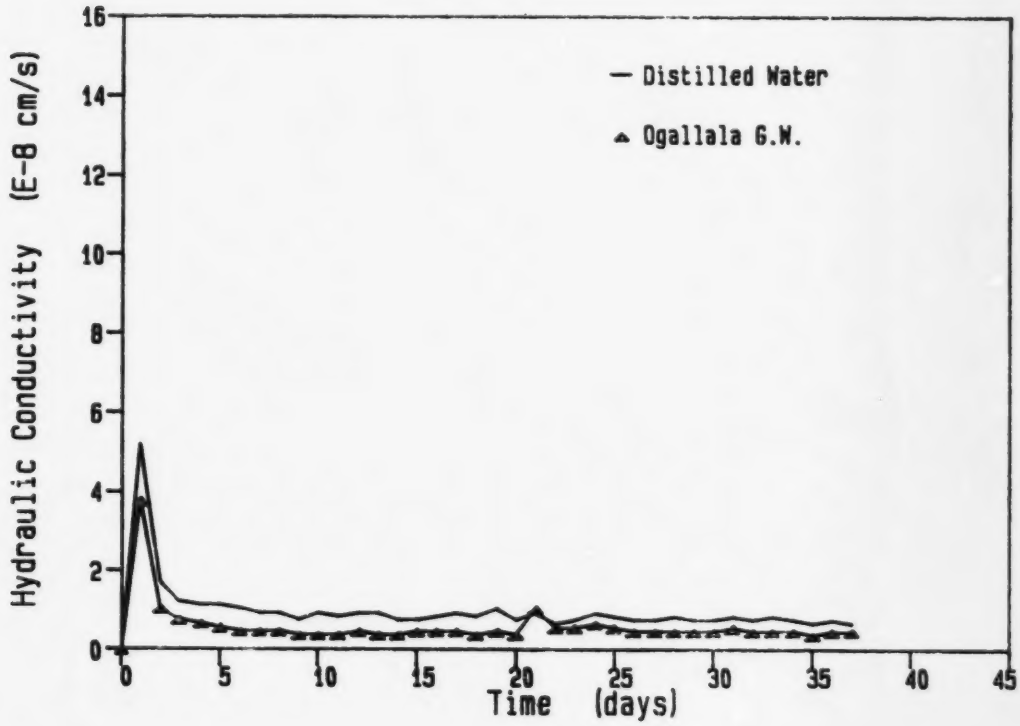

Figure A.12 Average hydraulic conductivity of 3 bentonite plugs determined by constant pressure injection with distilled water and with Ogallala groundwater at 138 kPa (20 psi). Hydraulic conductivity multiplier = 10^{-8} cm/s.

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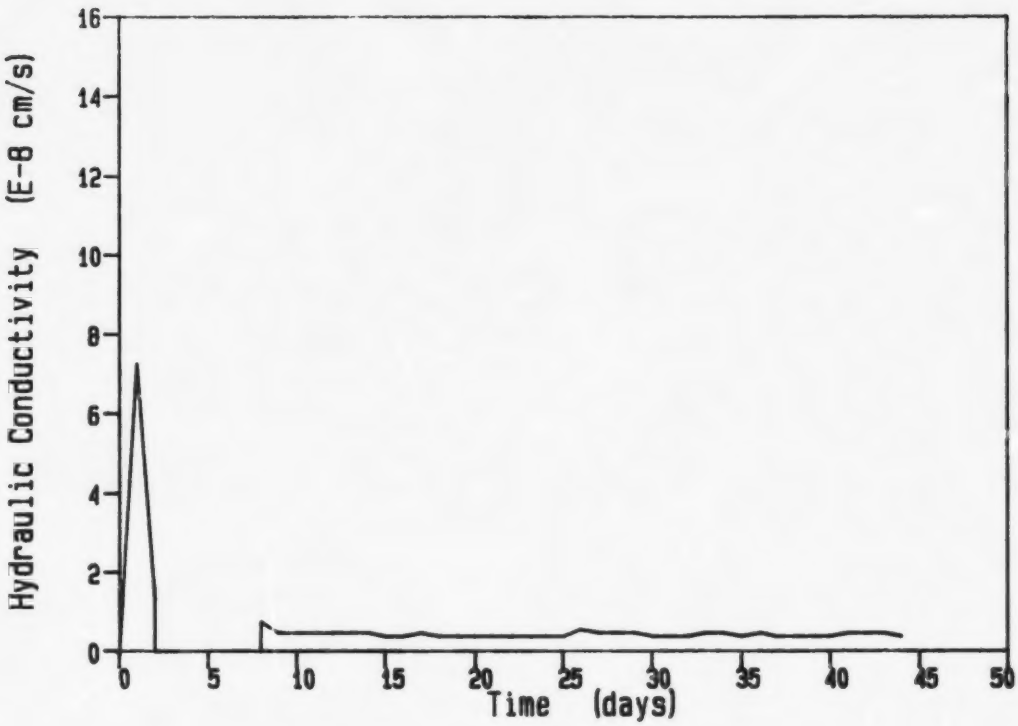

Figure A.13 Hydraulic conductivity of a 2.5 cm (1 inch) long bentonite plug with an initial moisture content of 75 percent. Hydraulic conductivity multiplier = 10^{-8} cm/s.

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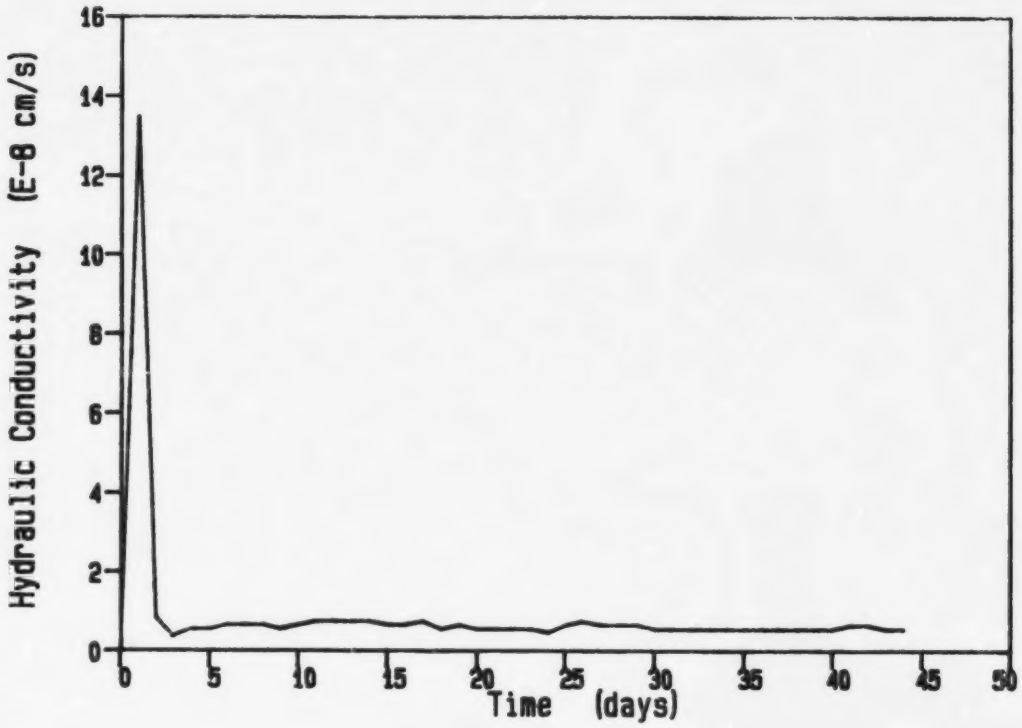

Figure A.14 Hydraulic conductivity of a 2.5 cm (1 inch) long bentonite plug with an initial moisture content of 100 percent. Hydraulic conductivity multiplier = 10^{-8} cm/s.

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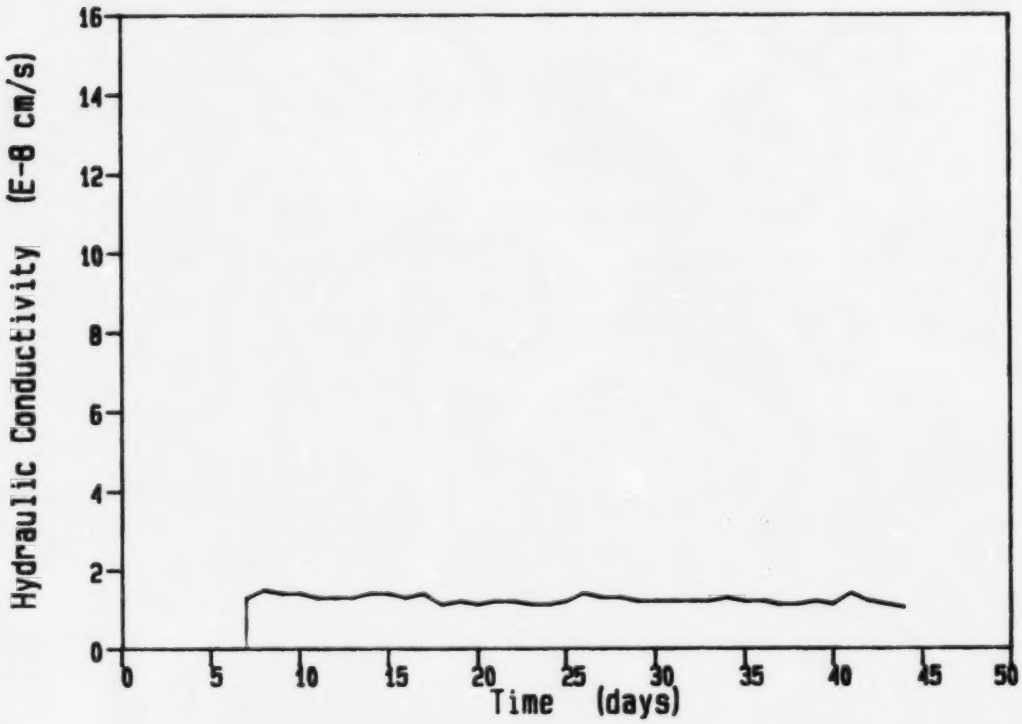

Figure A.15 Hydraulic conductivity of a 2.5 cm (1 inch) long bentonite plug with an initial moisture content of 125 percent. Hydraulic conductivity multiplier = 10^{-8} cm/s.

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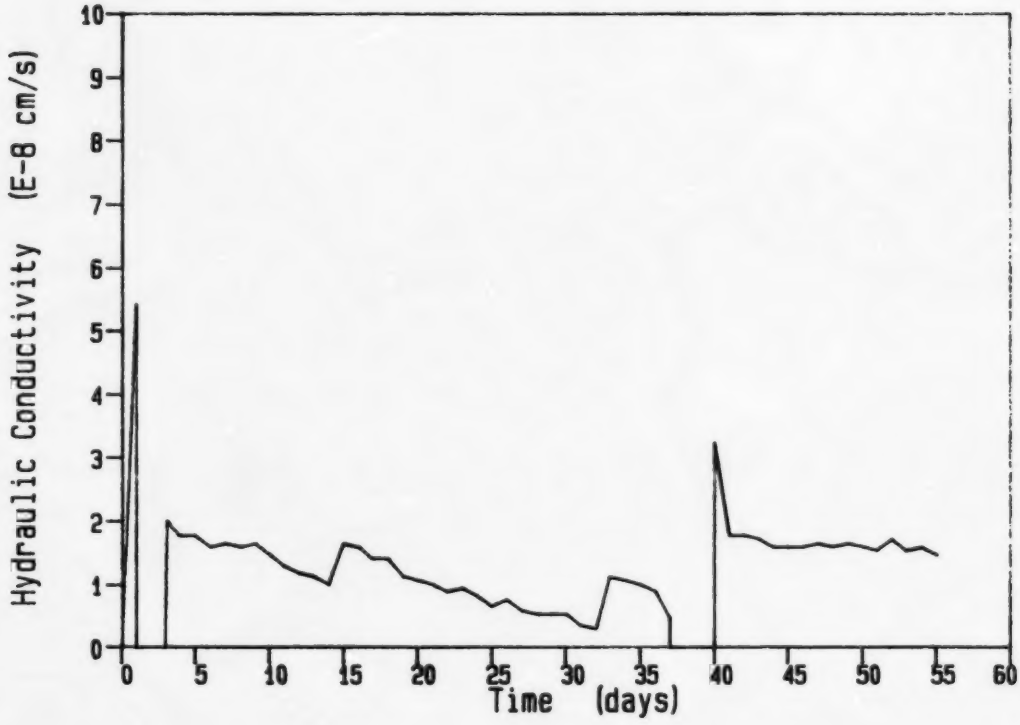

Figure A.16 Hydraulic conductivity of plug 1 determined by constant pressure injection testing using a 45 ppm calcium permeant. Hydraulic conductivity multiplier = 10^{-8} cm/s.

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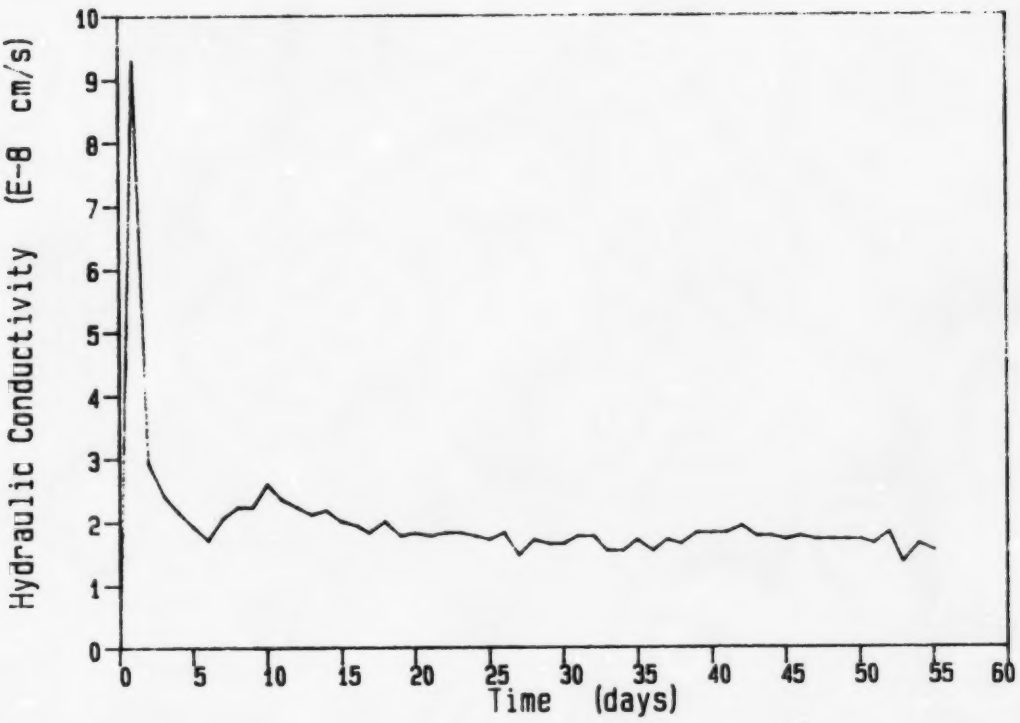

Figure A.17 Hydraulic conductivity of plug 2 determined by constant pressure injection-testing using a 45 ppm calcium permeant. Hydraulic conductivity multiplier = 10^{-8} cm/s.

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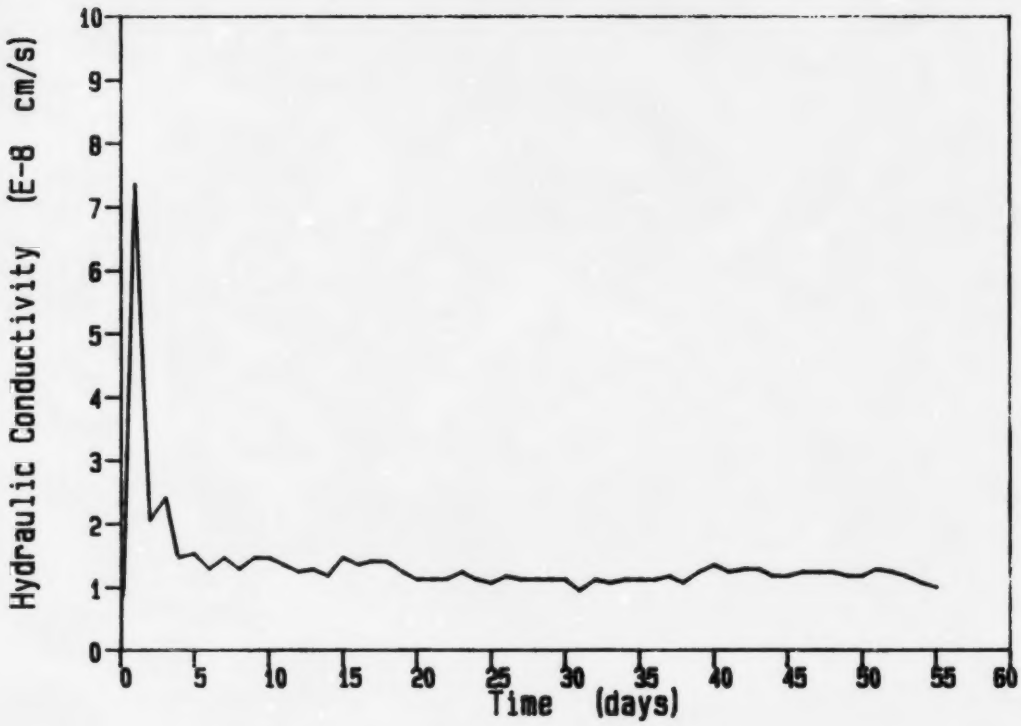

Figure A.18 Hydraulic conductivity of plug 3 determined by constant pressure injection testing using a 45 ppm calcium permeant. Hydraulic conductivity multiplier = 10^{-8} cm/s.

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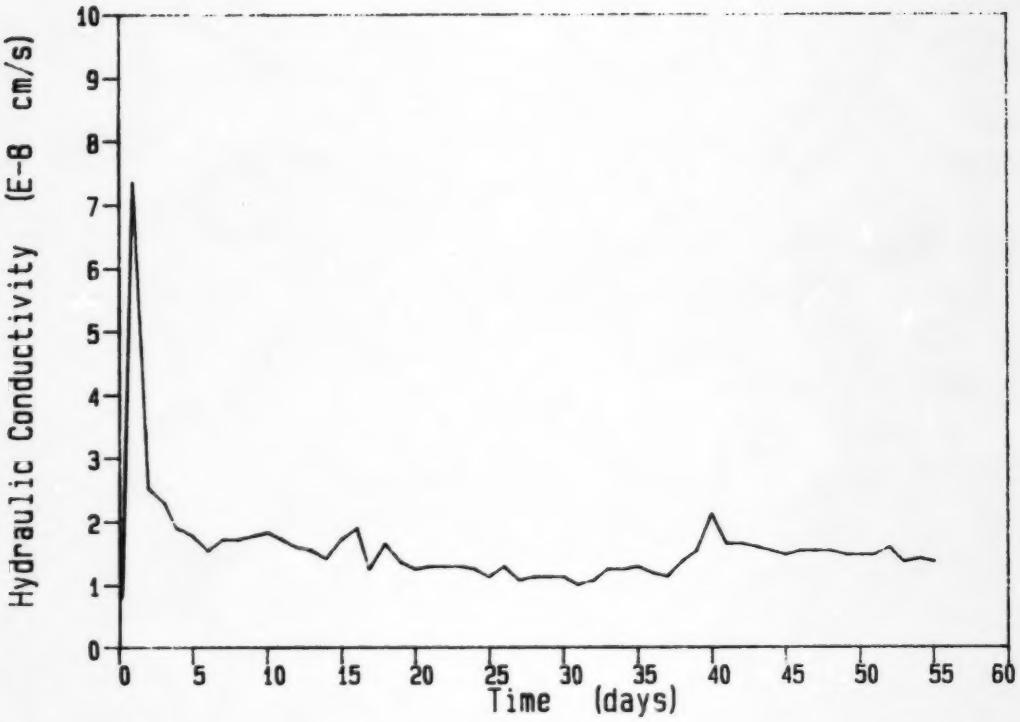

Figure A.19 Hydraulic conductivity of the 45 ppm calcium system determined by constant injection pressure testing. Hydraulic conductivity multiplier = 10^{-8} cm/s.

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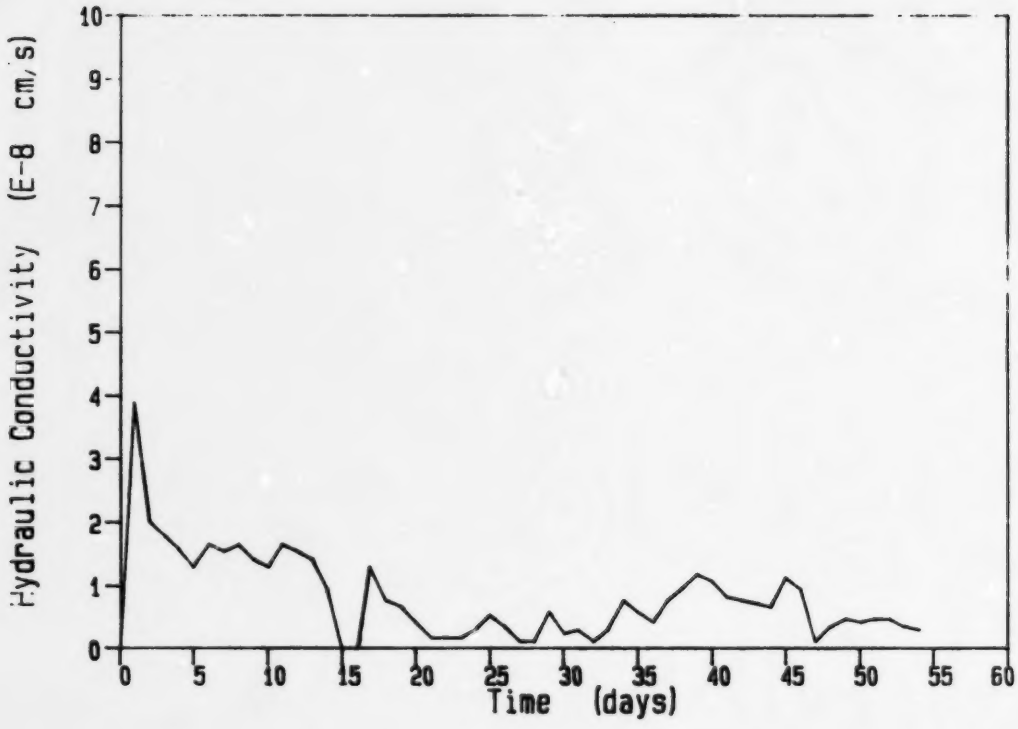

Figure A.20 Hydraulic conductivity of plug 1 determined by constant pressure injection testing using a 39 ppm sodium permeant. Small leak in collection system (Section 6.2.5). Hydraulic conductivity probably underestimated. Hydraulic conductivity multiplier = 10^{-8} cm/s.

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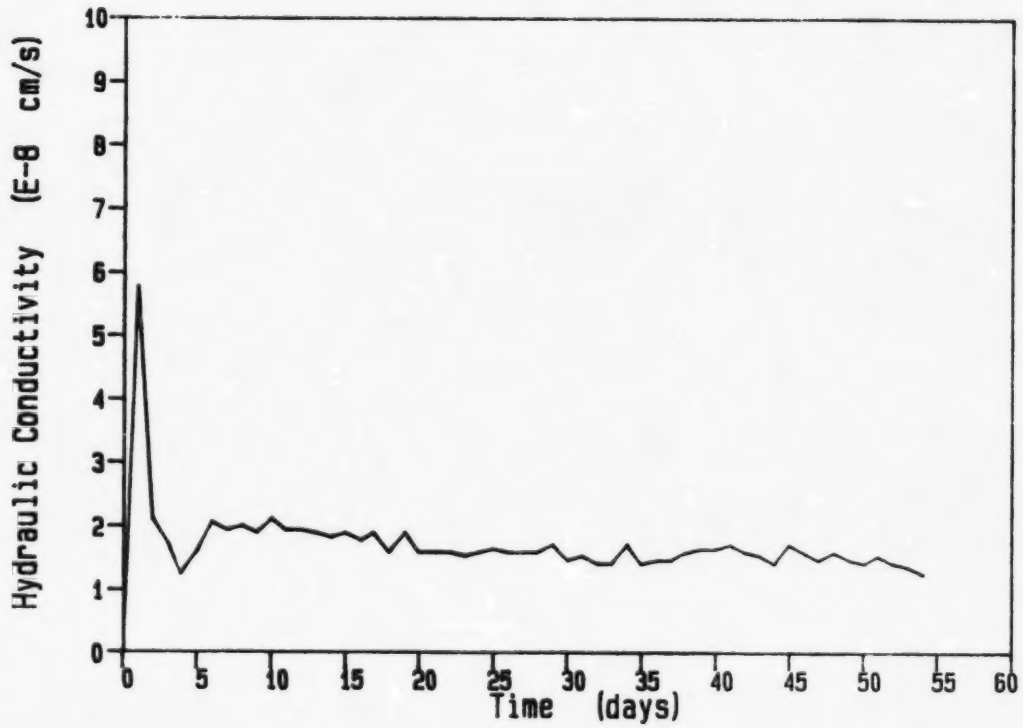

Figure A.21 Hydraulic conductivity of plug 3 determined by constant pressure injection testing using a 39 ppm sodium permeant. Hydraulic conductivity multiplier = 10^{-8} cm/s.

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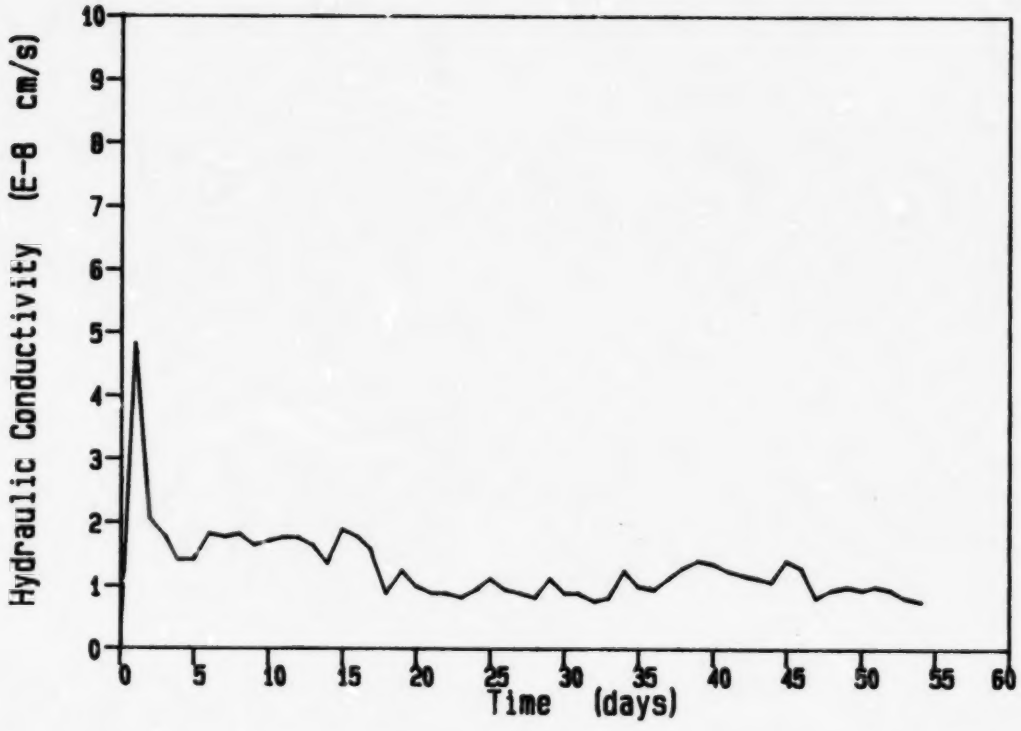

Figure A.22 Hydraulic conductivity of the 39 ppm sodium system (two plugs) determined by constant injection pressure testing. Hydraulic conductivity multiplier = 10^{-8} cm/s.

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APPENDIX B

MASS BALANCE PLOTS OBTAINED FROM CONSTANT PRESSURE INJECTION TESTING OF BENTONITE PLUGS

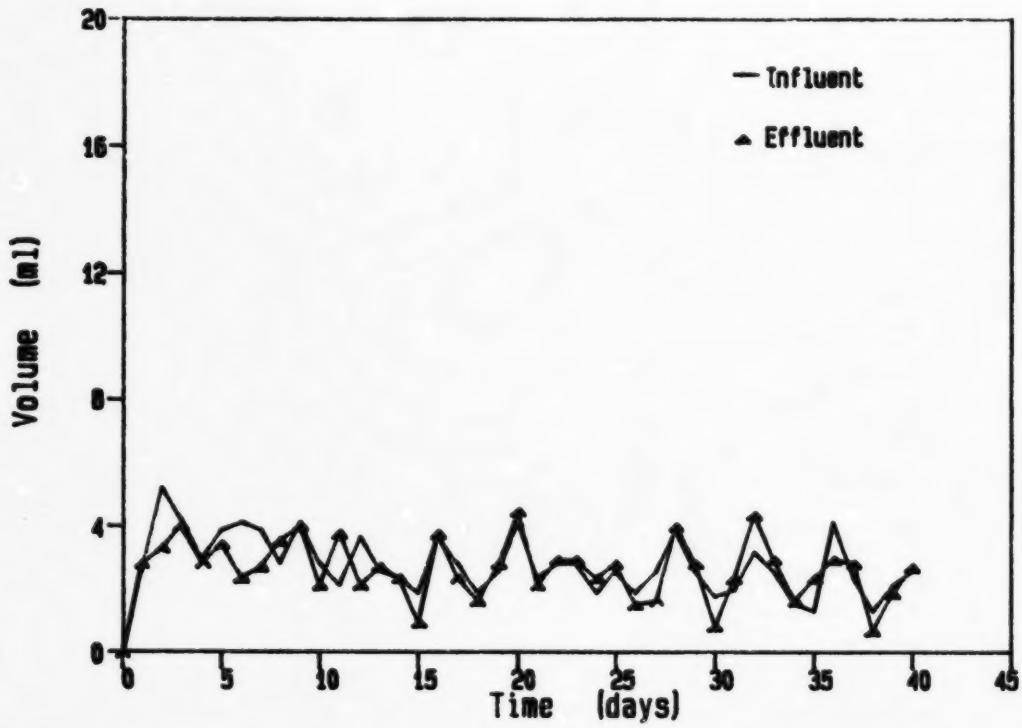

Figure B.1 Mass balance for constant pressure injection of plug 1 with distilled water at 34 kPa (5 psi).

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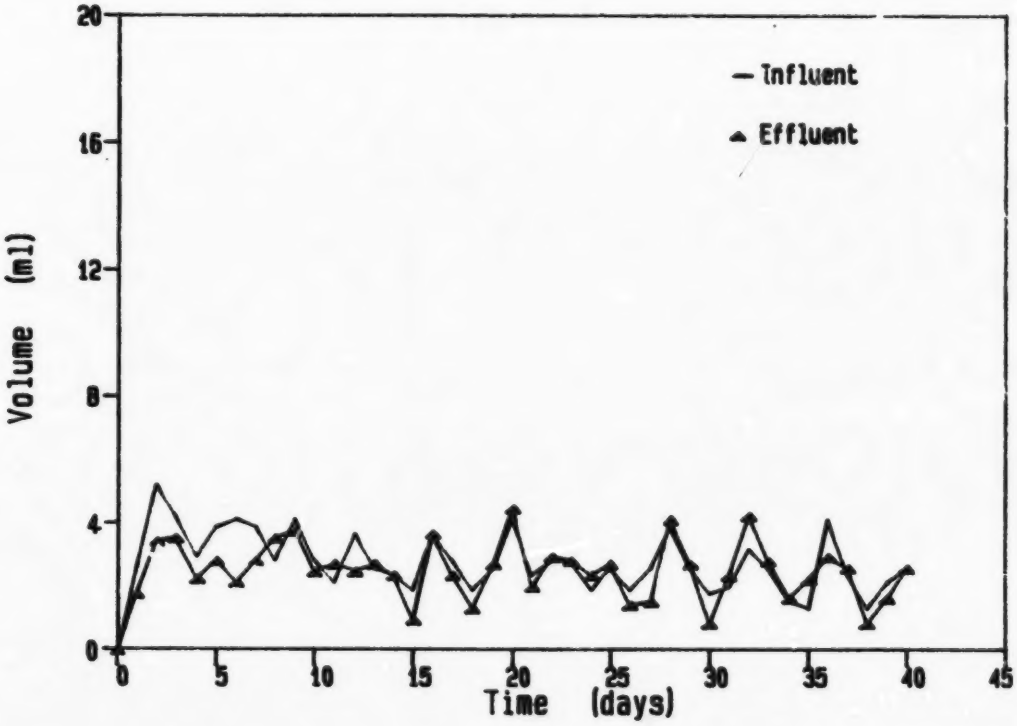

Figure B.2 Mass balance for constant pressure injection of plug 2 with distilled water at 34 kPa (5 psi) injection pressure.

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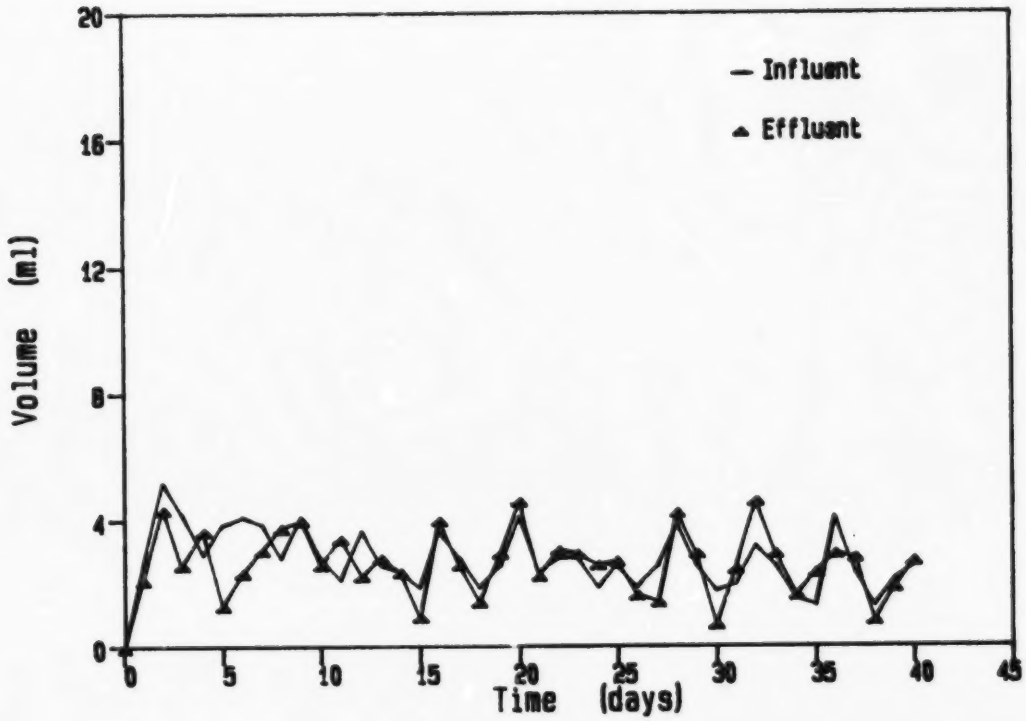

Figure B.3 Mass balance for constant pressure injection of plug 3 with distilled water at 34 kPa (5 psi).

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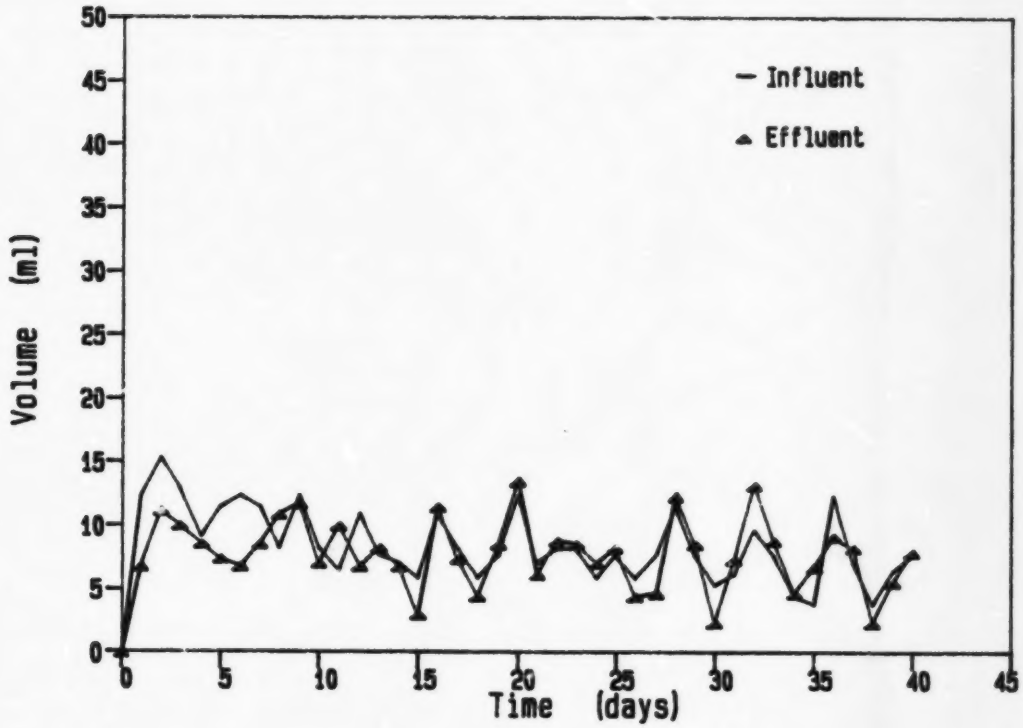

Figure B.4 Total mass balance (3 plugs) for constant pressure injection with distilled water at 34 kPa (5 psi).

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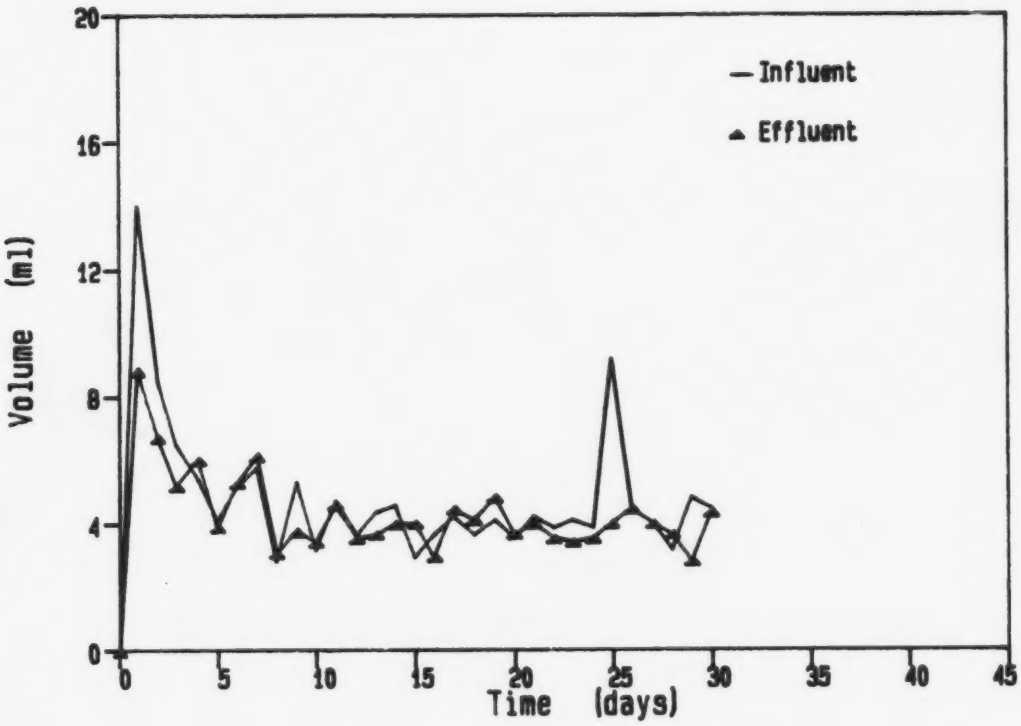

Figure B.5 Mass balance for constant pressure injection of plug 1 with distilled water at 69 kPa (10 psi).

Peak at 25 days probably is due to experimental error (Section 6.2.1).

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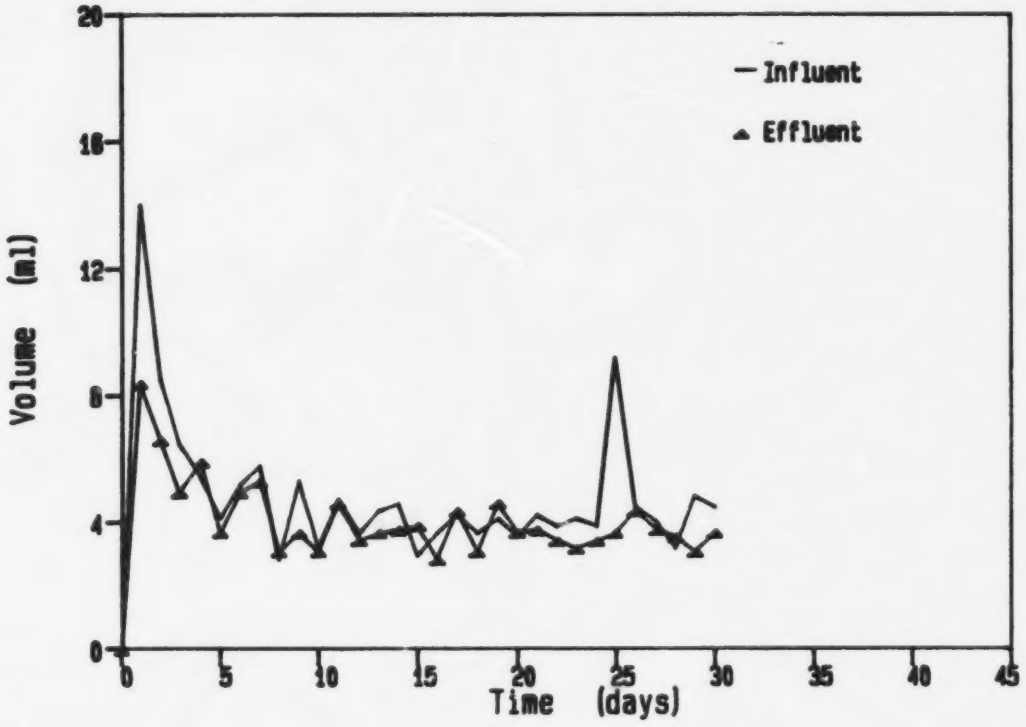

Figure B.6 Mass balance for constant pressure injection of plug 2 with distilled water at 69 kPa (10 psi).

Peak at 25 days probably is due to experimental error (Section 6.2.1).

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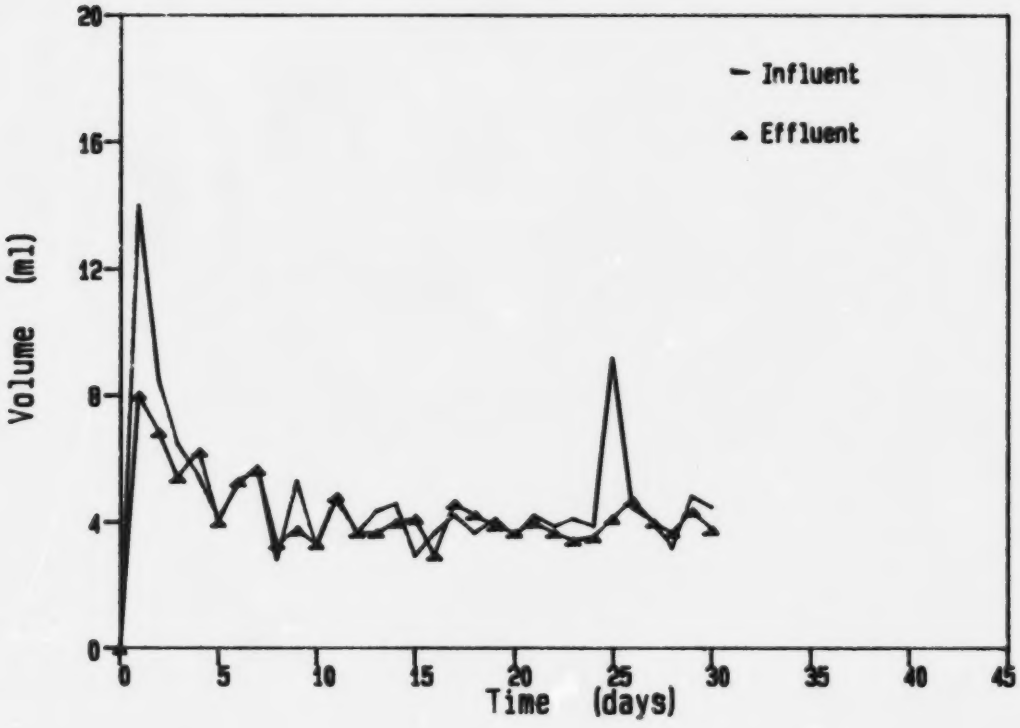

Figure B.7 Mass balance for constant pressure injection of plug 3 with distilled water at 69 kPa (10 psi).

Peak at 25 days probably is due to experimental error (Section 6.2.1).

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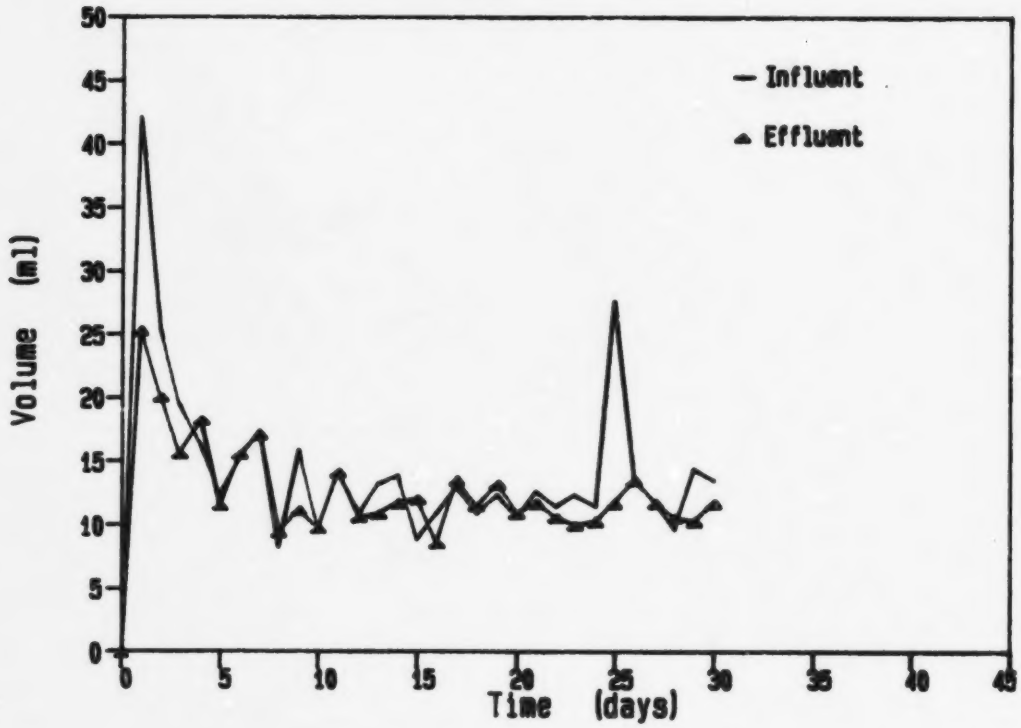

Figure B.8 Total mass balance (3 plugs) for constant pressure injection with distilled water at 69 kPa (10 psi).

Peak at 25 days probably is due to experimental error (Section 6.2.1).

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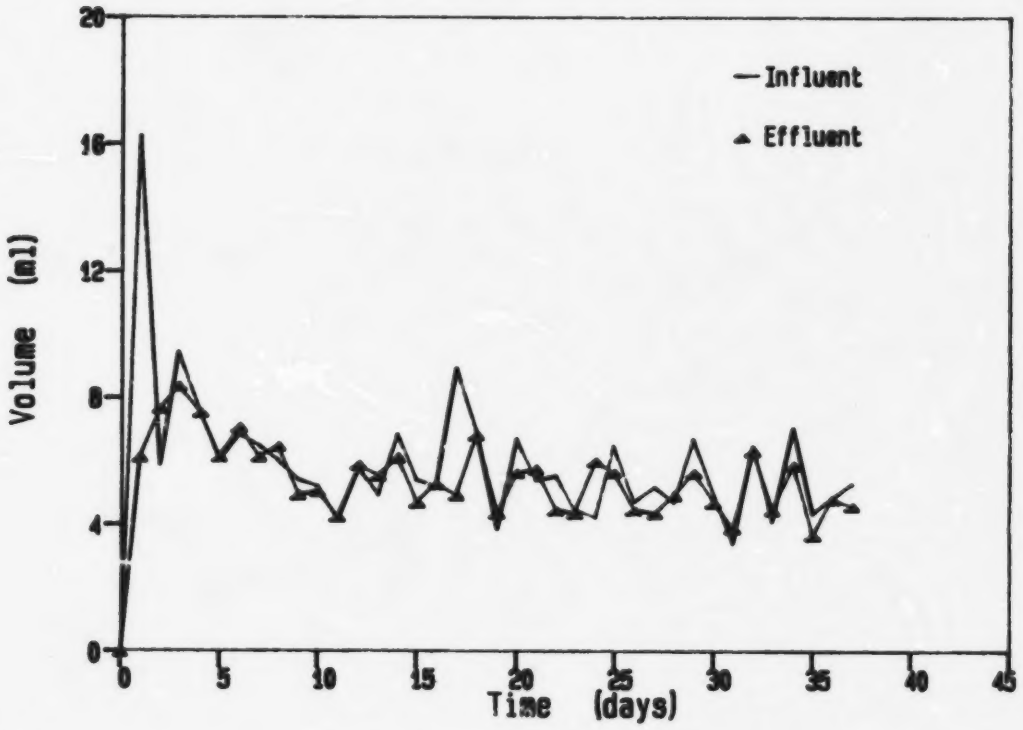

Figure B.9 Mass balance for constant pressure injection of plug 1 with distilled water at 138 kPa (20 psi).

Peak at 17 days is probably due to experimental error (Section 6.2.1).

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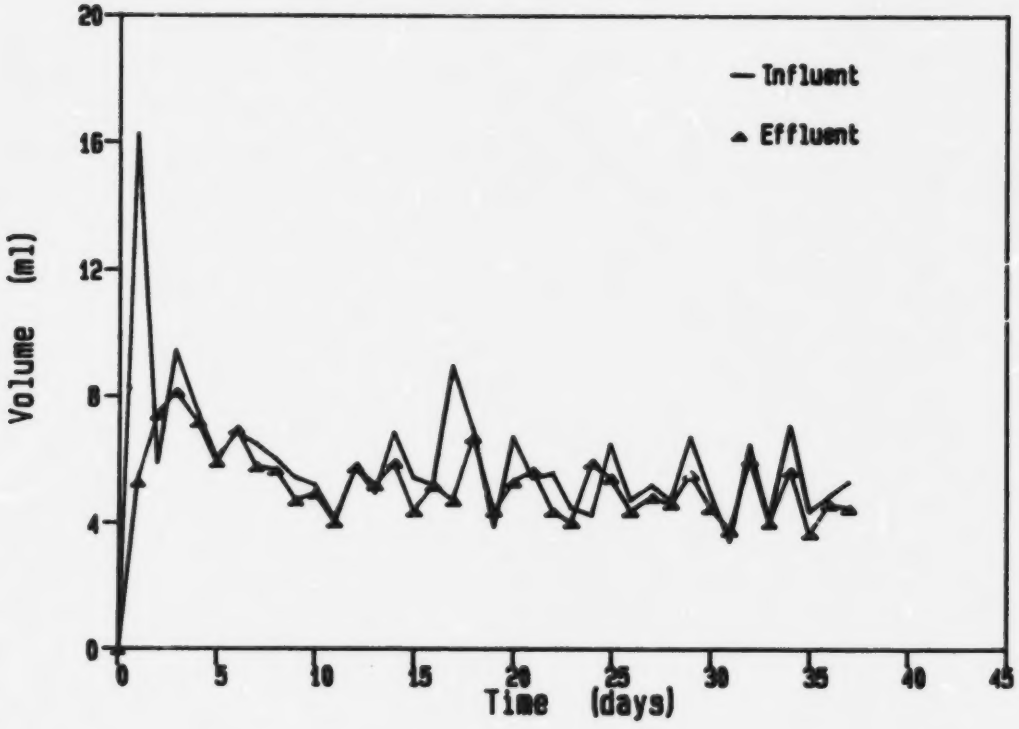

Figure B.10 Mass balance for constant pressure injection of plug 2 with distilled water at 138 kPa (20 psi).

Peak at 17 days probably is due to experimental error (Section 6.2.1).

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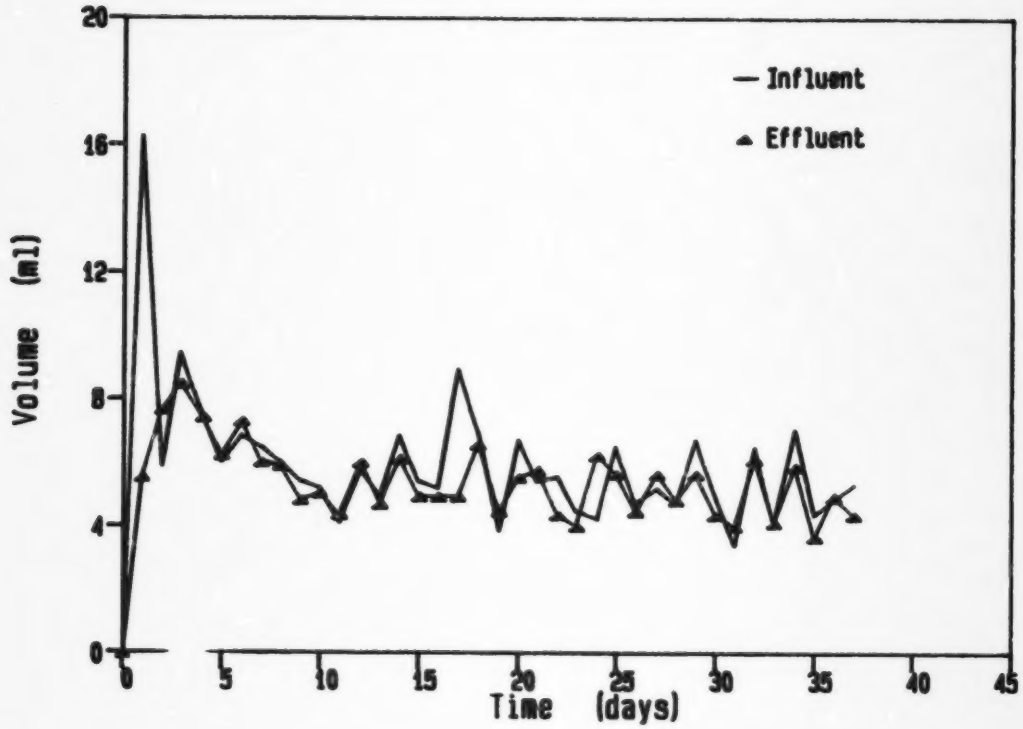

Figure B.11 Mass balance for constant pressure injection of plug 3 with distilled water at 138 kPa (20 psi).

Peak at 17 days probably is due to experimental error (Section 6.2.1).

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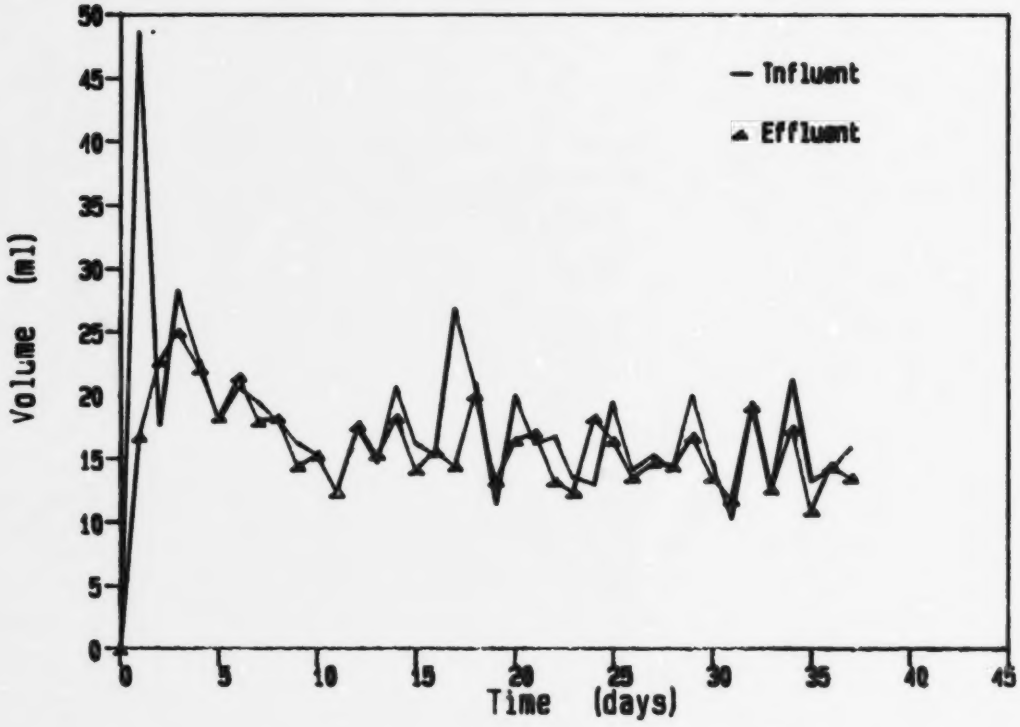

Figure B.12 Total mass balance (3 plugs) for constant pressure injection with distilled water at 138 kPa (20 psi).

Peak at 17 days probably is due to experimental error (Section 6.2.1).

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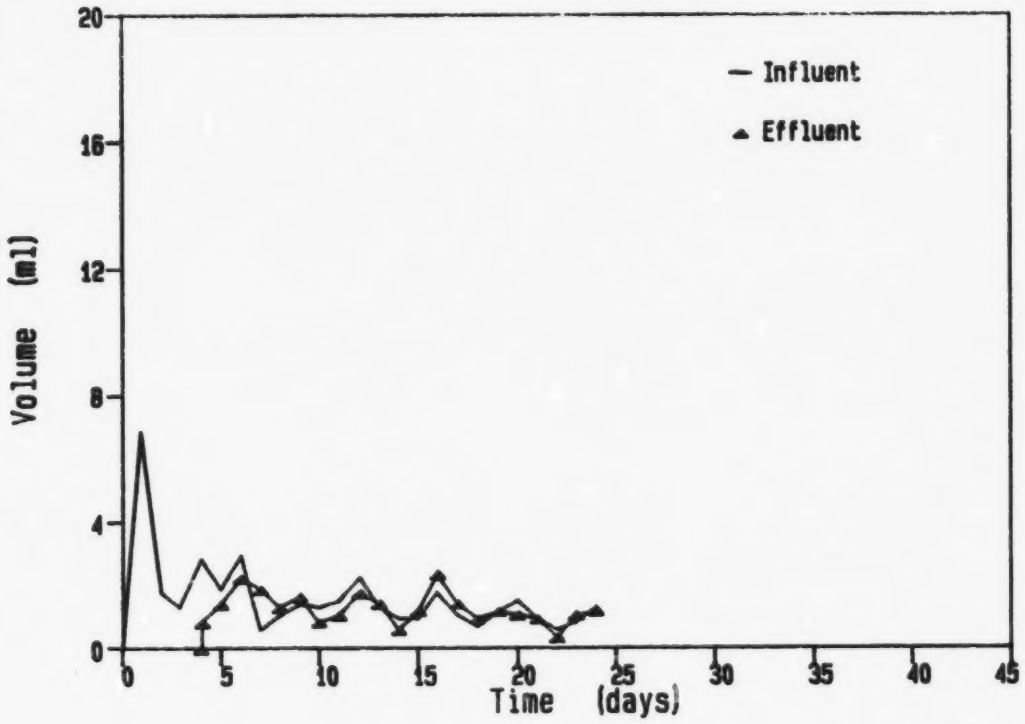

Figure B.13 Mass balance for constant pressure flow testing of plug 1 using Ogallala groundwater at 34 kPa (5 psi) injection pressure.

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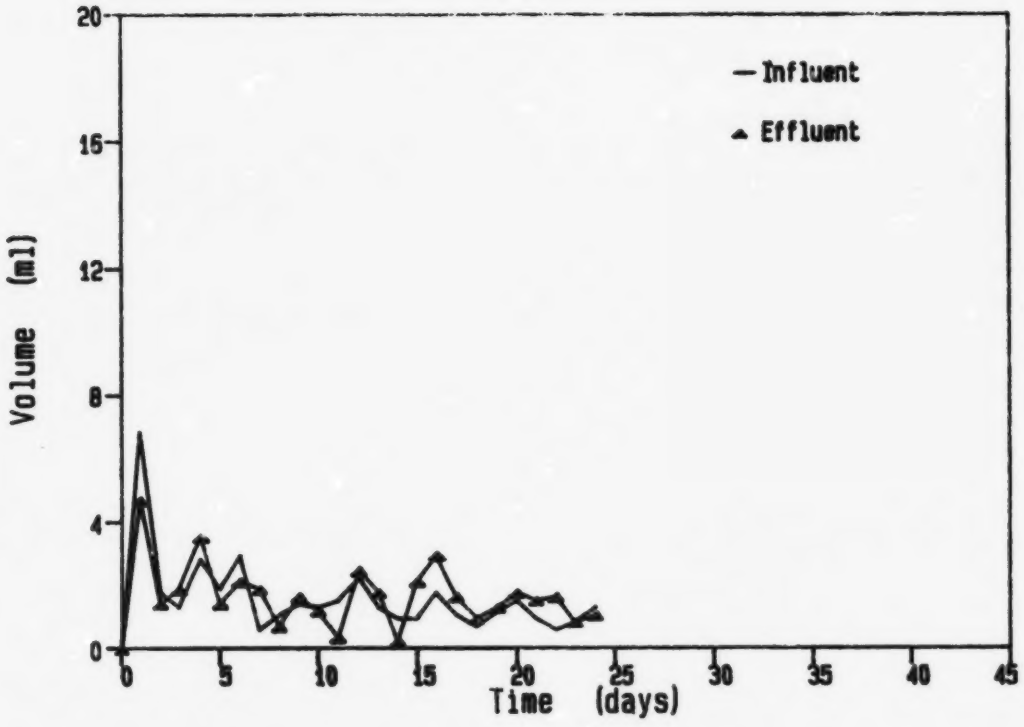

Figure B.14 Mass balance for constant pressure flow testing of plug 2 using Ogallala groundwater at 34 kPa (5 psi) injection pressure.

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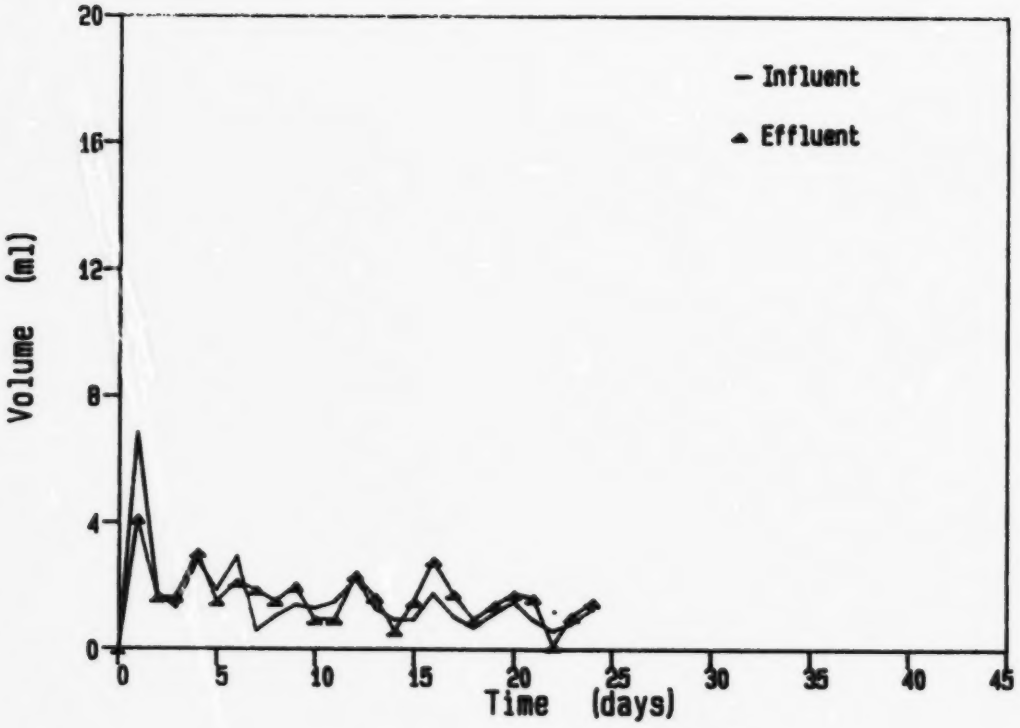

Figure B.15 Mass balance for constant pressure flow testing of plug 3 using Ogallala groundwater at 34 kPa (5 psi) injection pressure.

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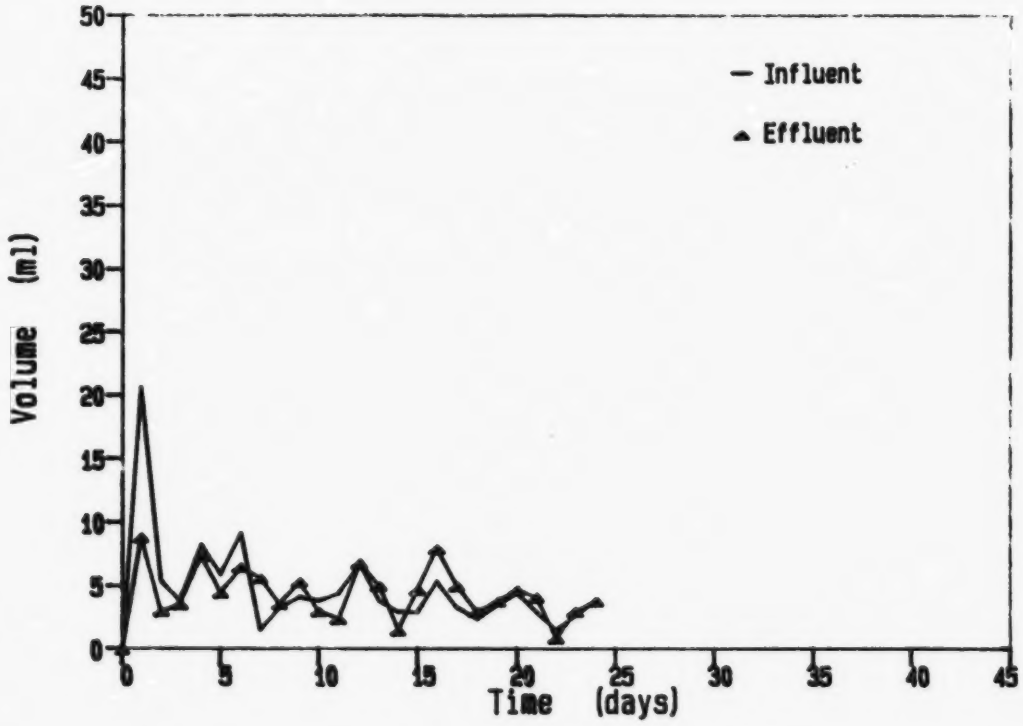

Figure B.16 Total mass balance (3 plugs) for constant pressure flow testing of the Ogallala system at 34 kPa (5 psi) injection pressure.

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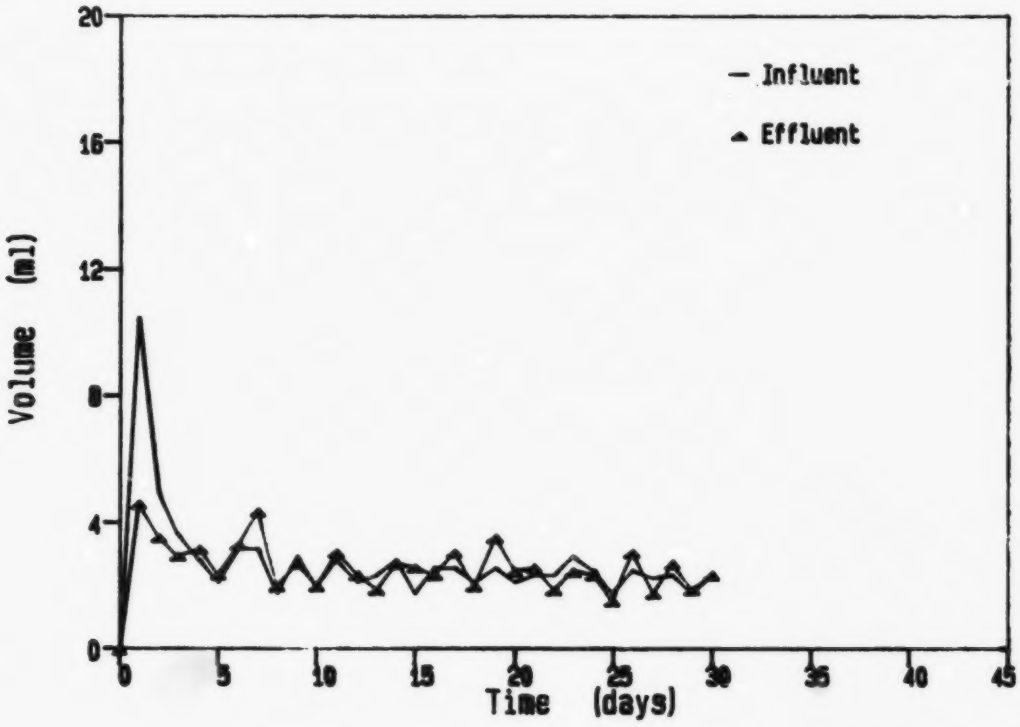

Figure B.17 Mass balance for constant pressure flow testing of plug 1 using Ogallala groundwater at 69 kPa (10 psi) injection pressure.

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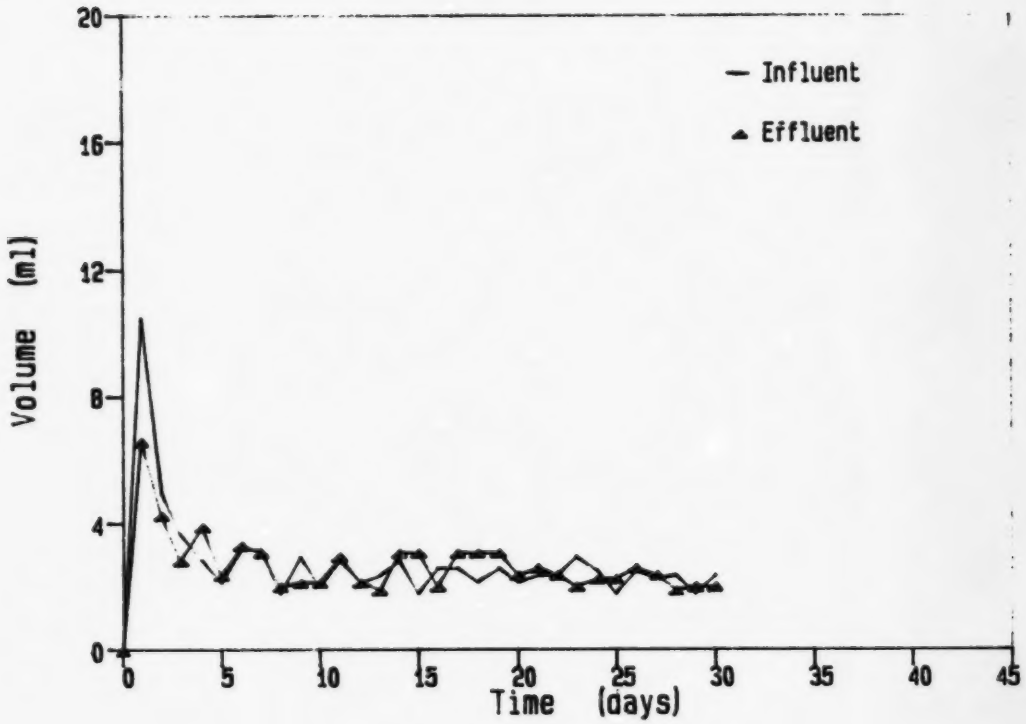

Figure B.18 Mass balance for constant pressure flow testing of plug 2 using Ogallala groundwater at 69 kPa (10 psi) injection pressure.

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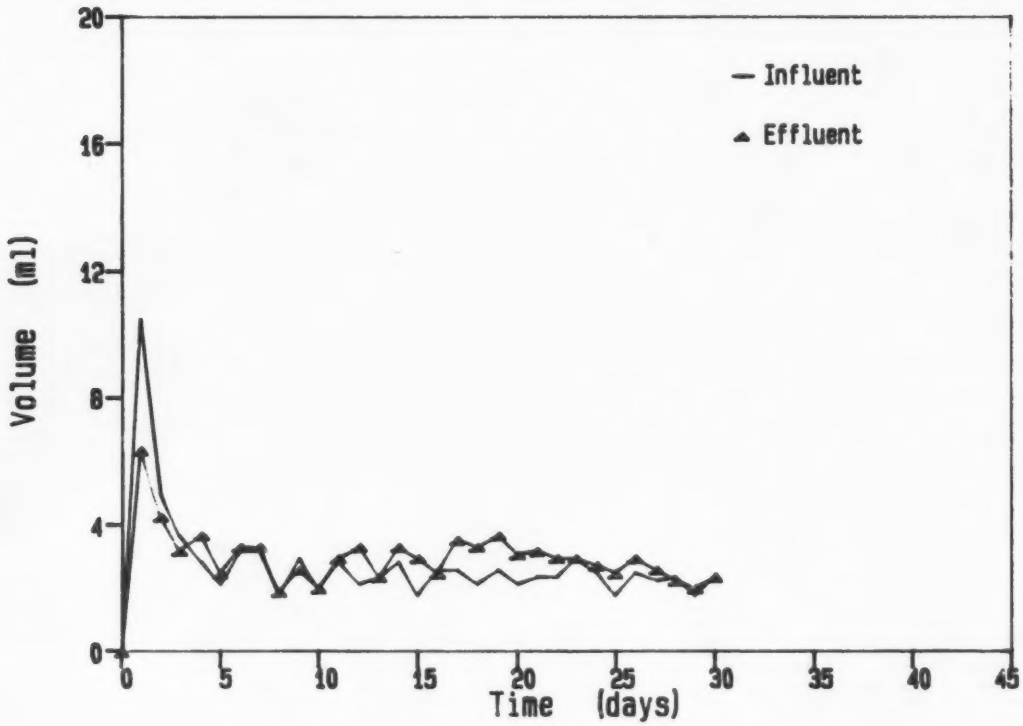

Figure B.19 Mass balance for constant pressure flow testing of plug 3 using Ogallala groundwater at 69 kPa (10 psi) injection pressure.

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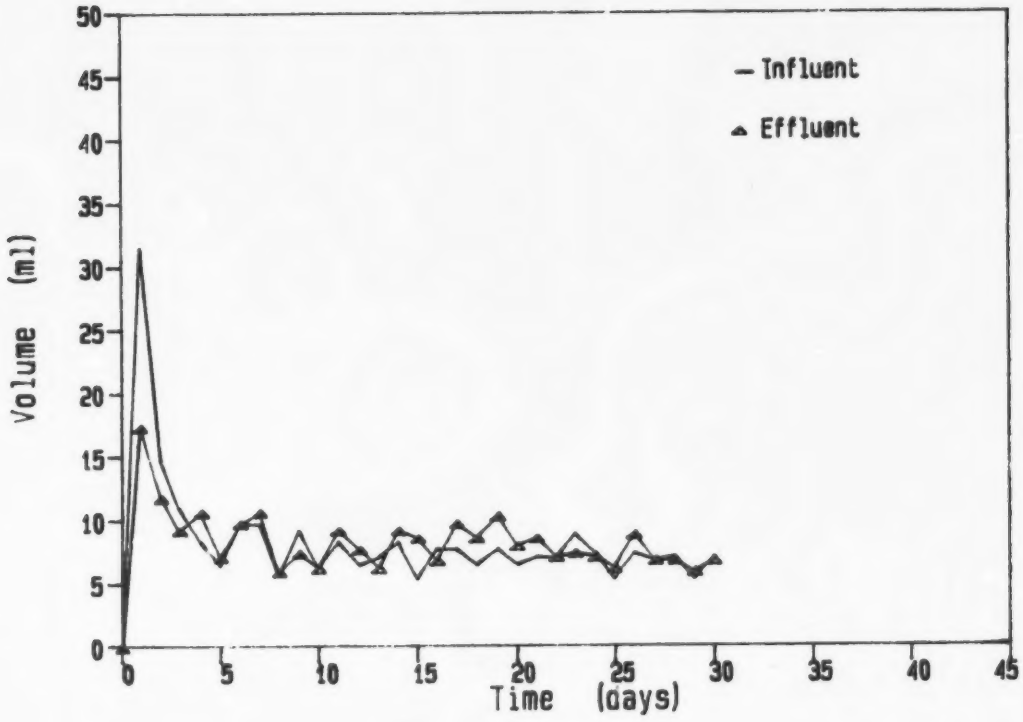

Figure B.20 Total mass balance (3 plugs) for constant pressure flow testing of the Ogallala system at 69 kPa (10 psi) injection pressure.

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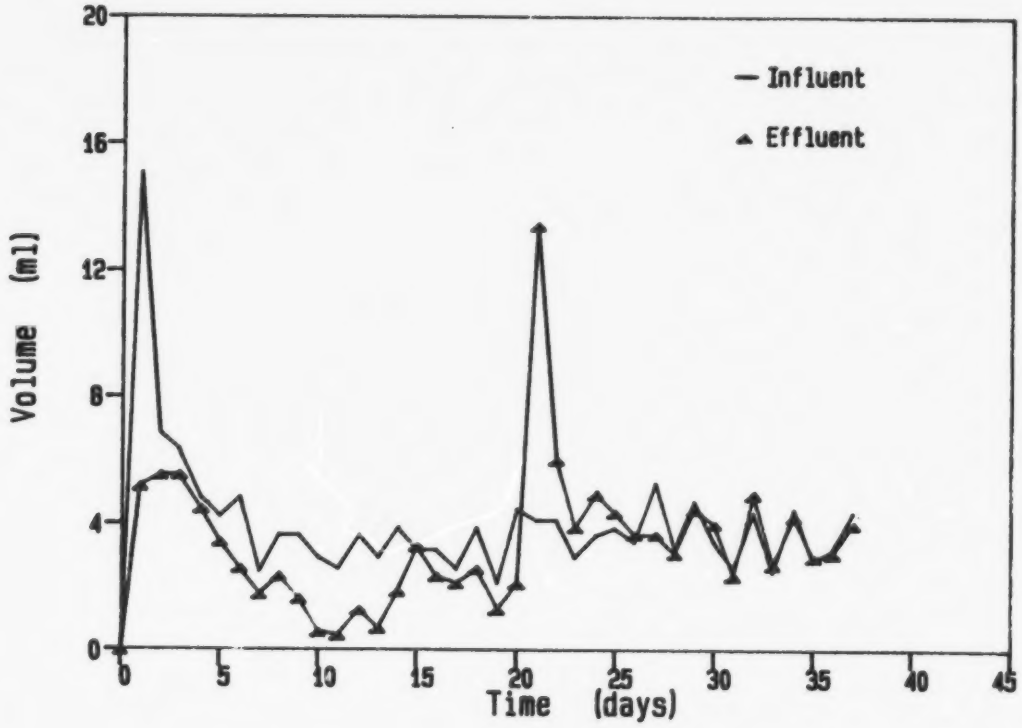

Figure B.21 Mass balance for constant pressure flow testing of plug 1 using Ogallala groundwater at 138 kPa (20 psi) injection pressure.

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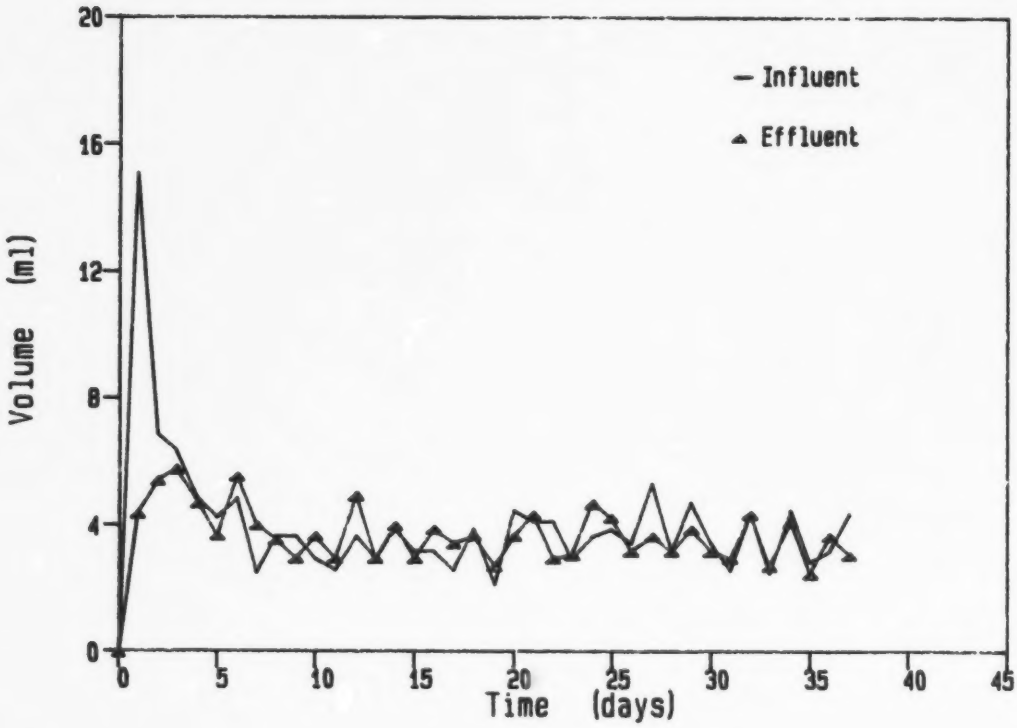

Figure B.22 Mass balance for constant pressure flow testing of plug 2 using Ogallala groundwater at 138 kPa (20 psi) injection pressure.

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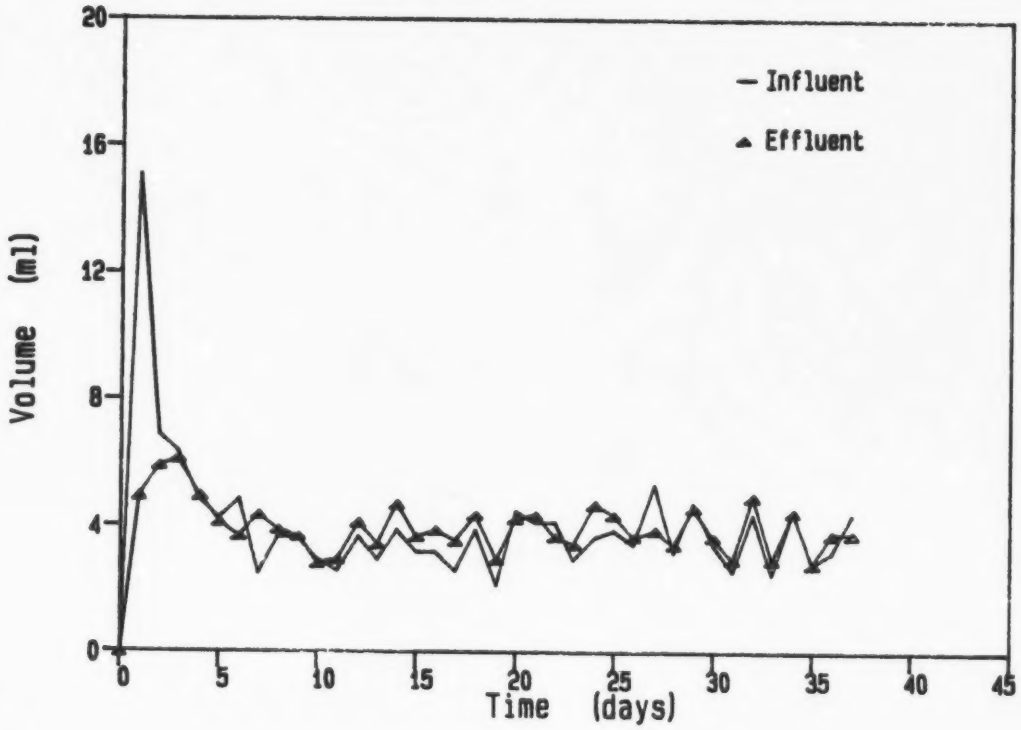

Figure B.23 Mass balance for constant pressure flow testing of plug 3 using Ogallala groundwater at 138 kPa (20 psi) injection pressure.

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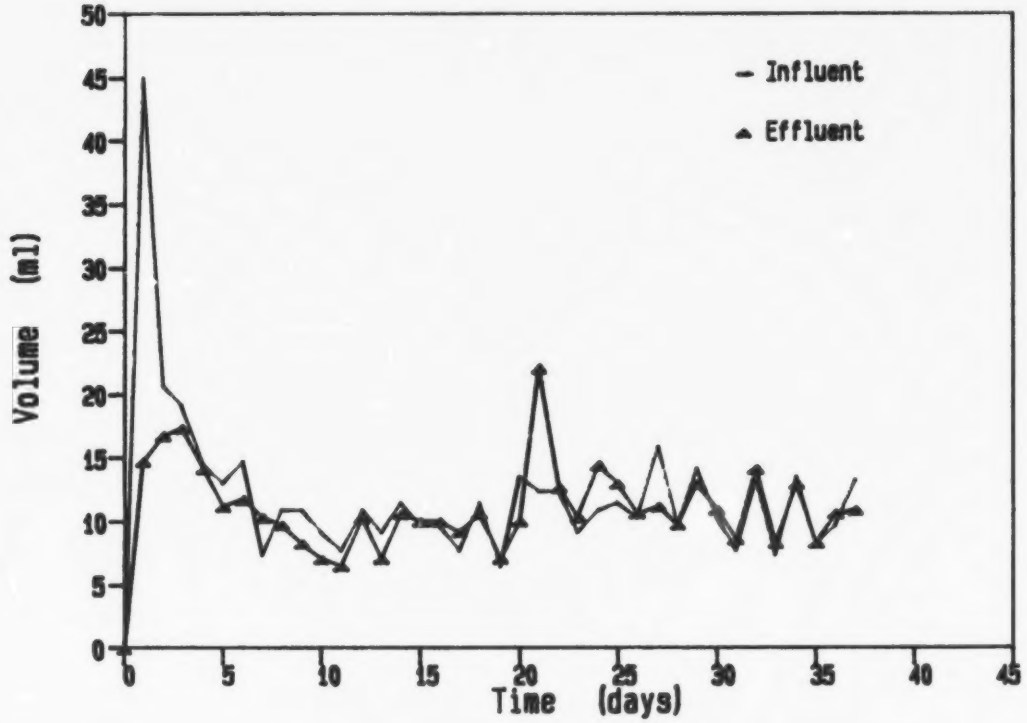

Figure B.24 Total mass balance (3 plugs) for constant pressure flow testing of the Ogallala system at 138 kPa (20 psi) injection pressure.

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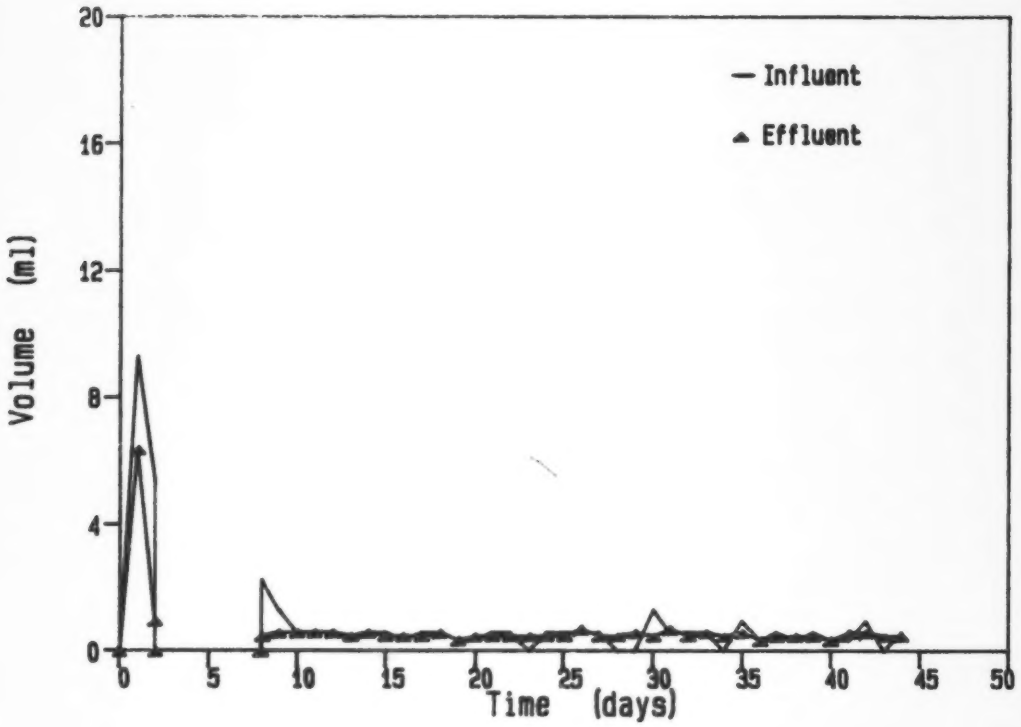

Figure B.25 Mass balance for constant pressure injection testing of a 2.5 cm (1 inch) long bentonite plug having an initial moisture content of 75 percent, using a 1000 ppm calcium permeant.

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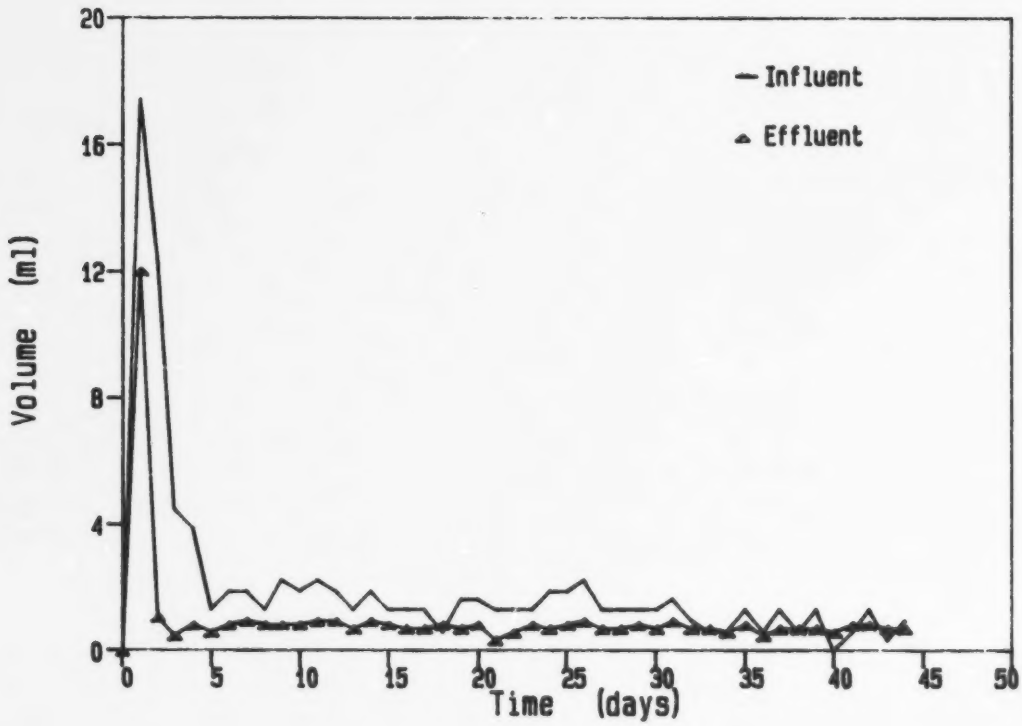

Figure B.26 Mass balance for constant pressure injection testing of a 2.5 cm (1 inch) long bentonite plug having an initial moisture content of 100 percent, using a 1000 ppm calcium permeant.

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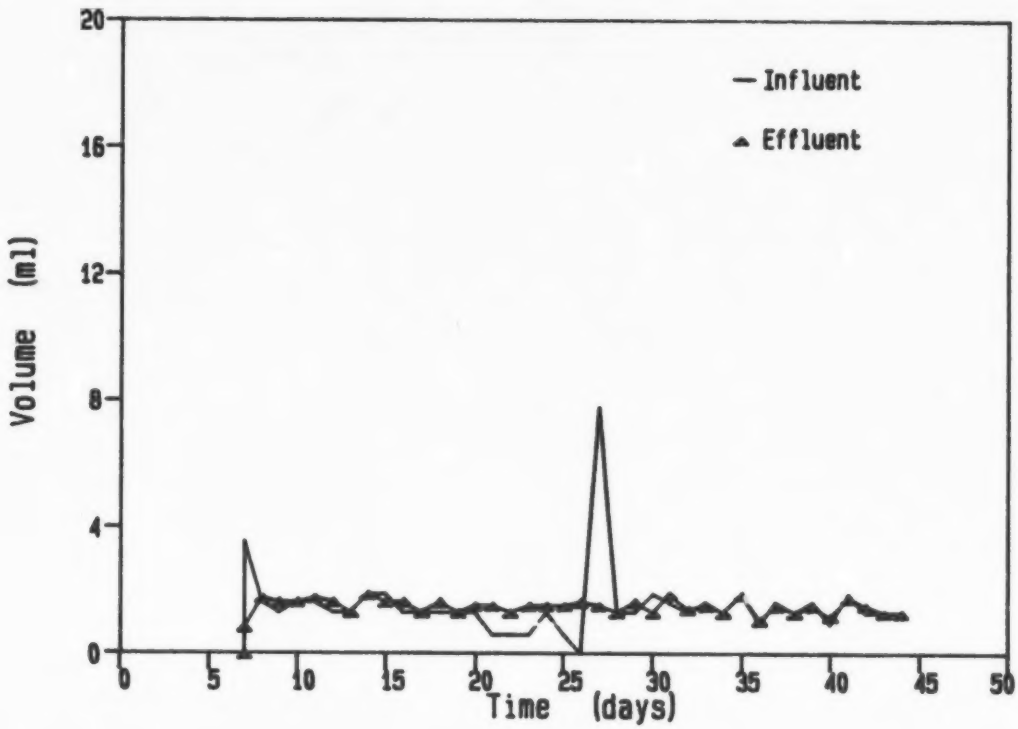

Figure B.27 Mass balance for constant pressure injection testing of a 2.5 cm (1 inch) long bentonite plug having an initial moisture content of 125 percent, using a 1000 ppm calcium permeant.

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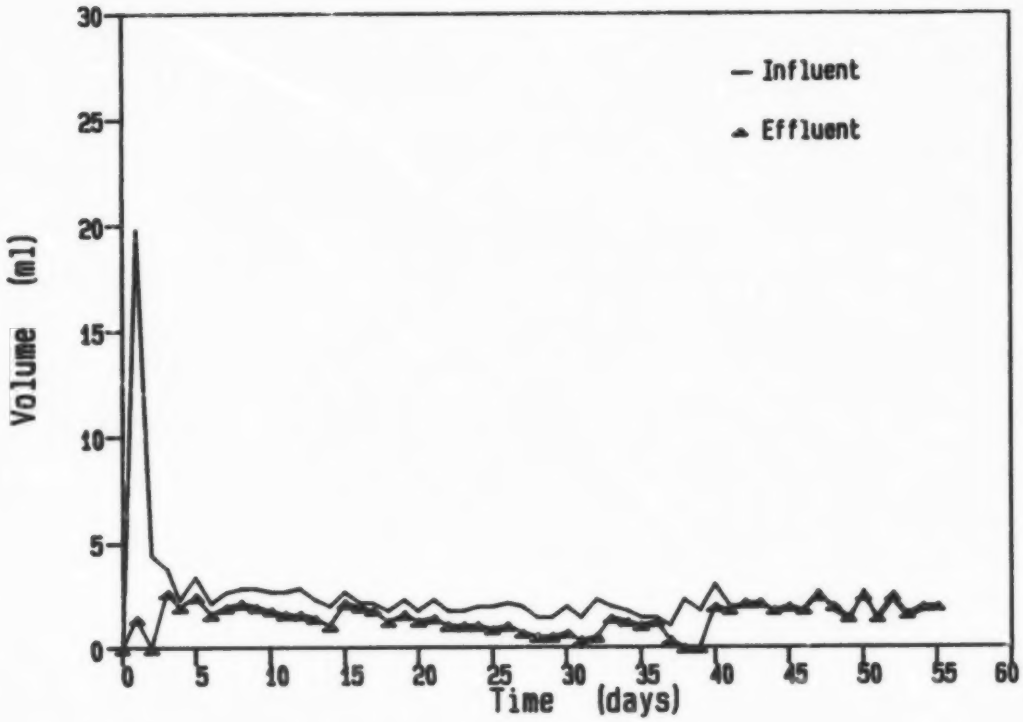

Figure B.28 Mass balance for constant pressure injection testing of plug 1 using a 45 ppm calcium permeant.

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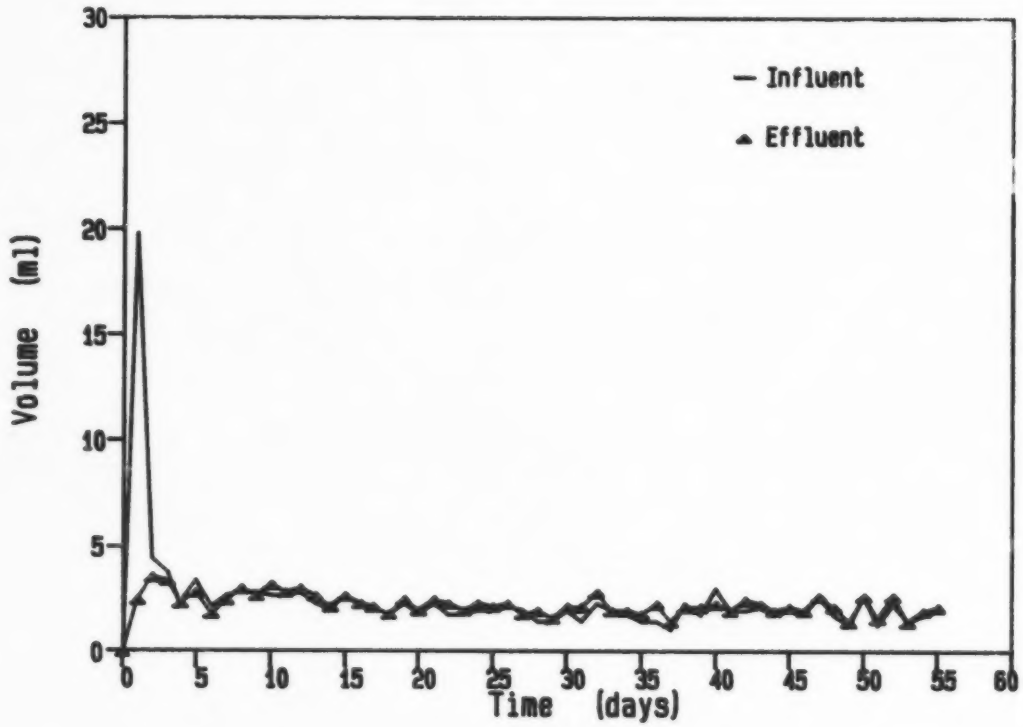

Figure B.29 Mass balance for constant pressure injection testing of plug 2 using a 45 ppm calcium permeant.

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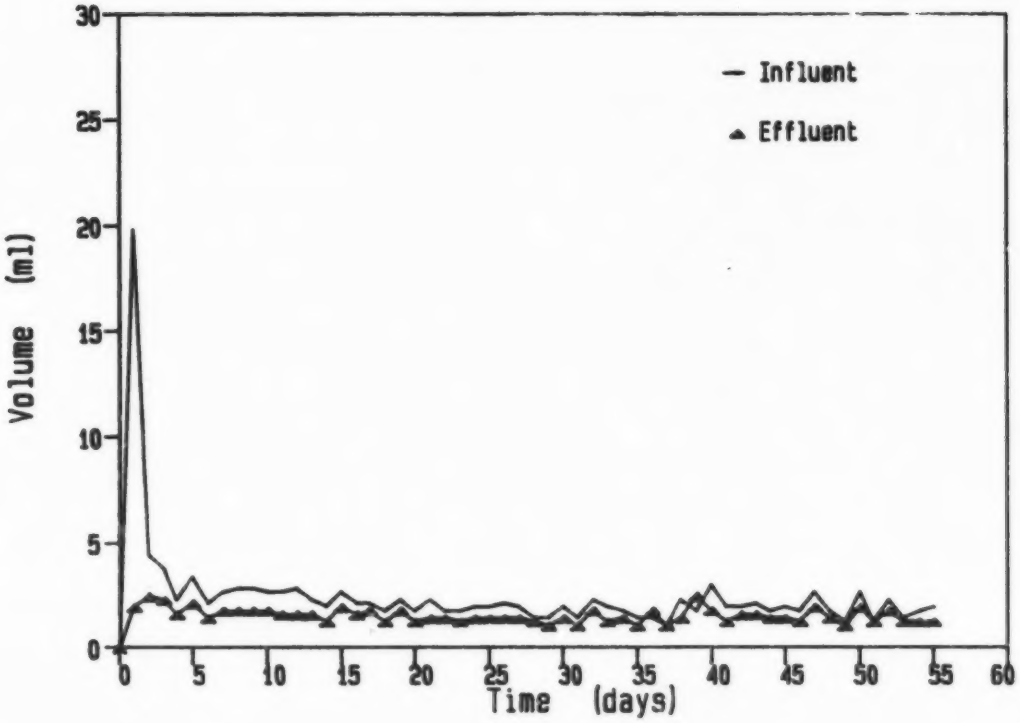

Figure B.30 Mass balance for constant pressure injection testing of plug 3 using a 45 ppm calcium permeant.

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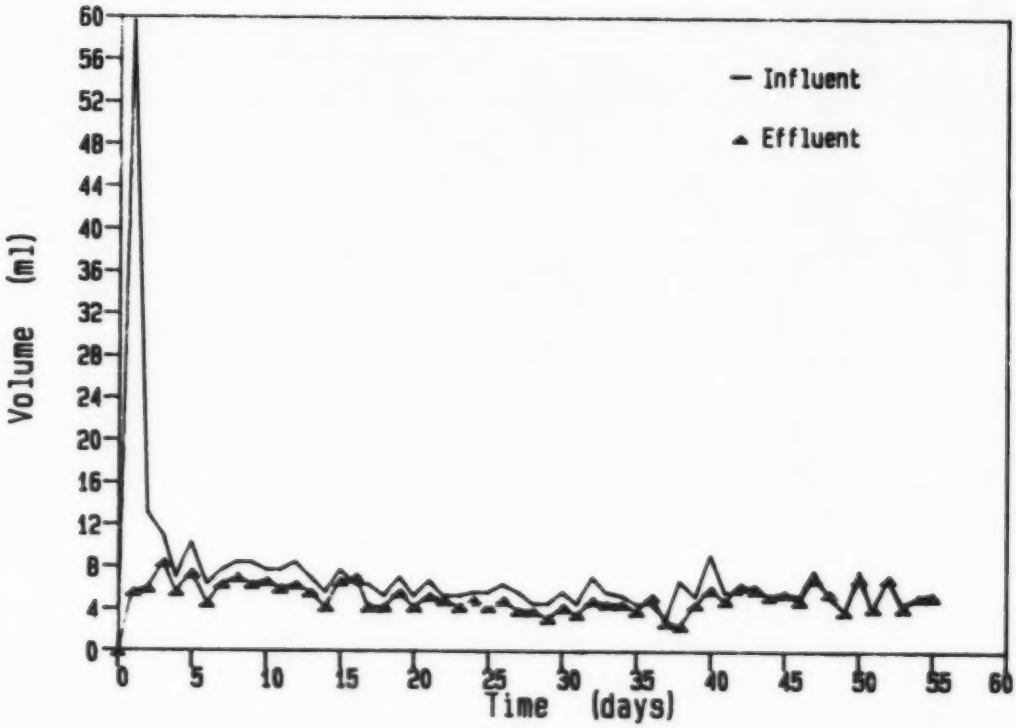

Figure B.31 Total mass balance for constant pressure injection testing of three 2.5 cm (1 inch) long bentonite plugs using a 45 ppm calcium permeant.

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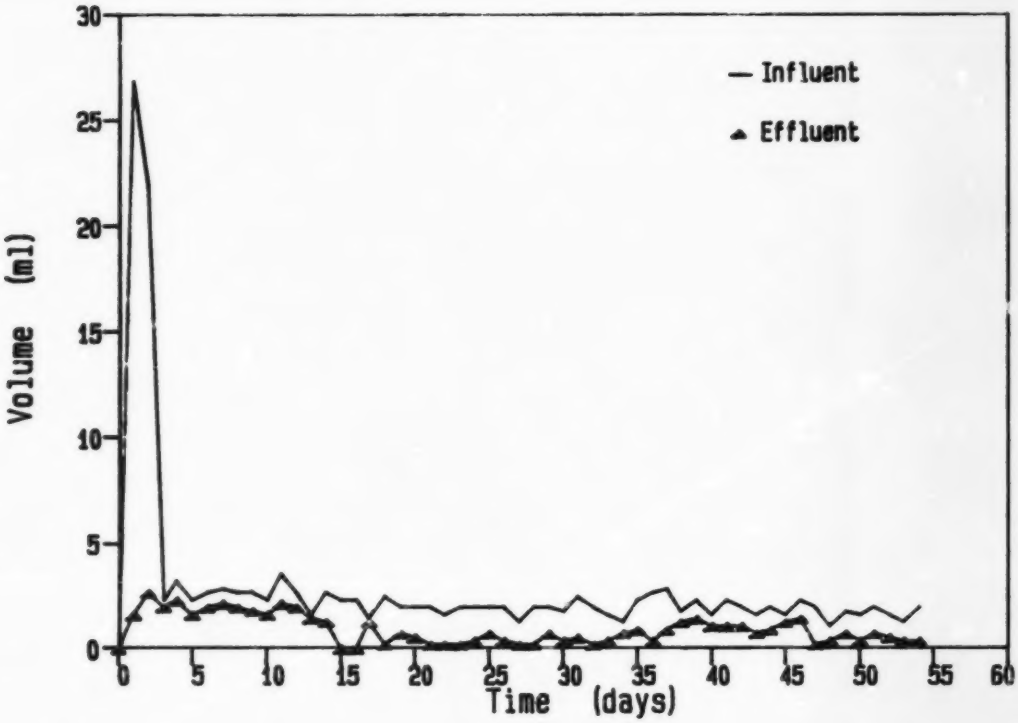

Figure B.32 Mass balance for constant pressure injection testing of plug 1 using a 39 ppm sodium permeant.

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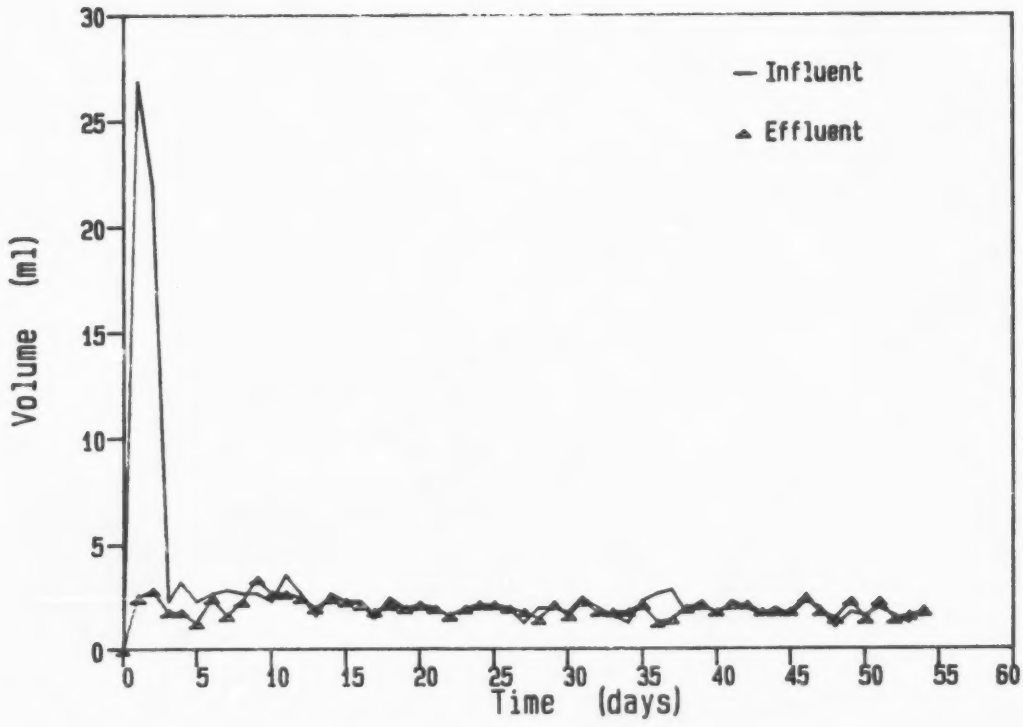

Figure B.33 Mass balance for constant pressure injection testing of plug 3 using a 39 ppm sodium permeant.

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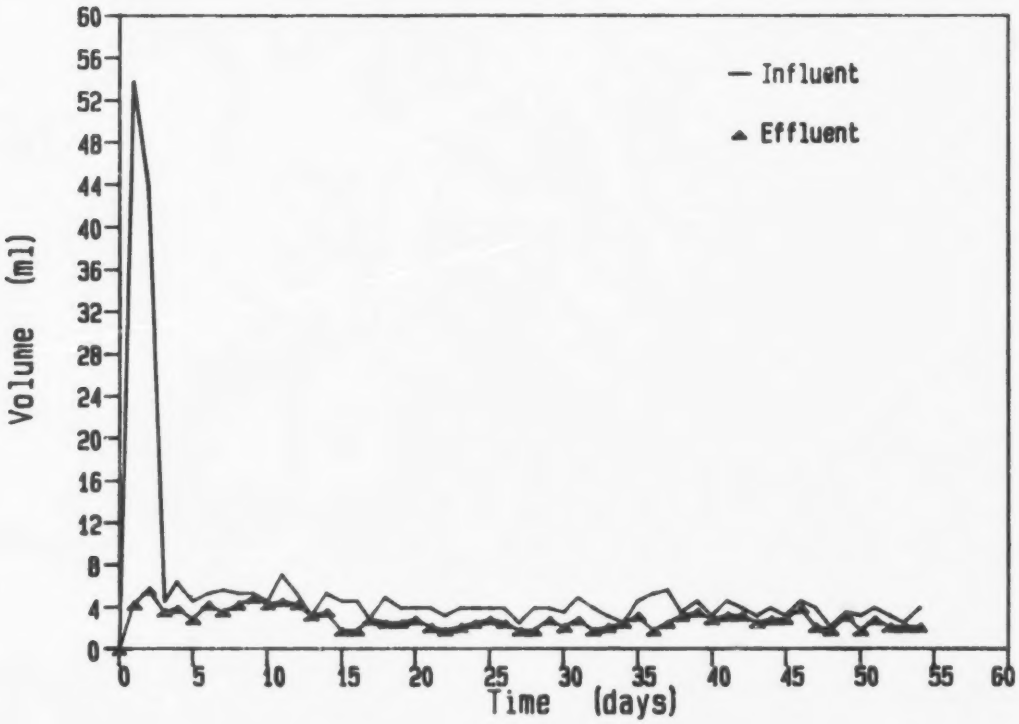

Figure B.34 Total mass balance for constant pressure injection testing of two 2.5 cm (1 inch) long bentonite plugs using a 39 ppm sodium permeant.

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APPENDIX C

CONSOLIDATION PLOTS OBTAINED FROM CONSOLIDATION
OF BENTONITE PLUGS USING NITROGEN

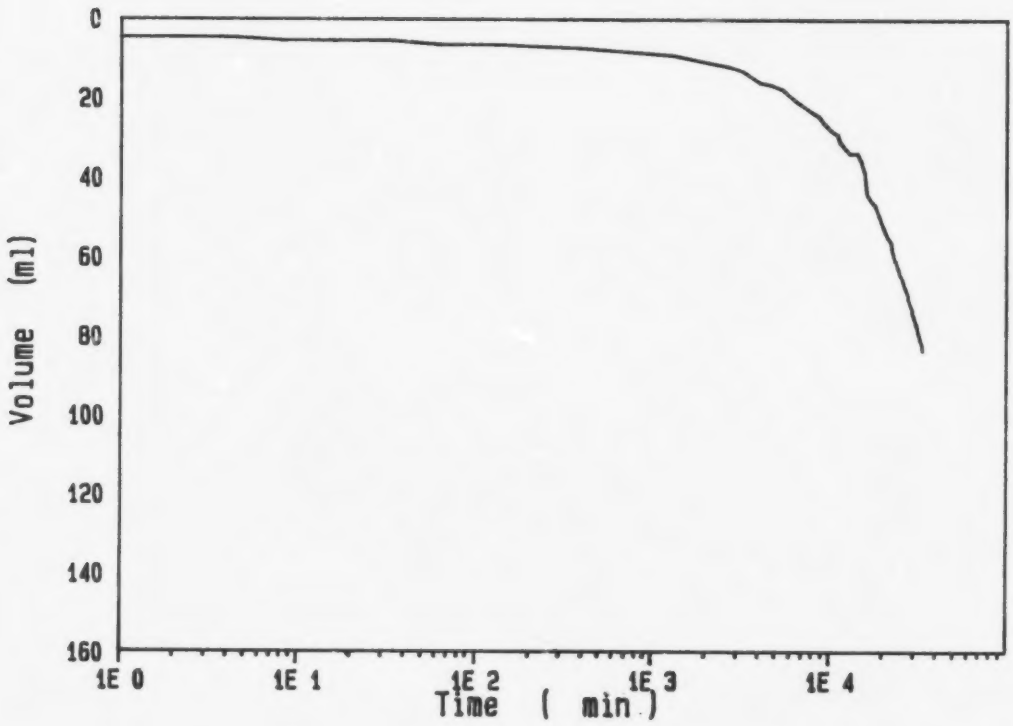

Figure C.1 Average consolidation for three bentonite plugs consolidated at 34 kPa (5 psi).

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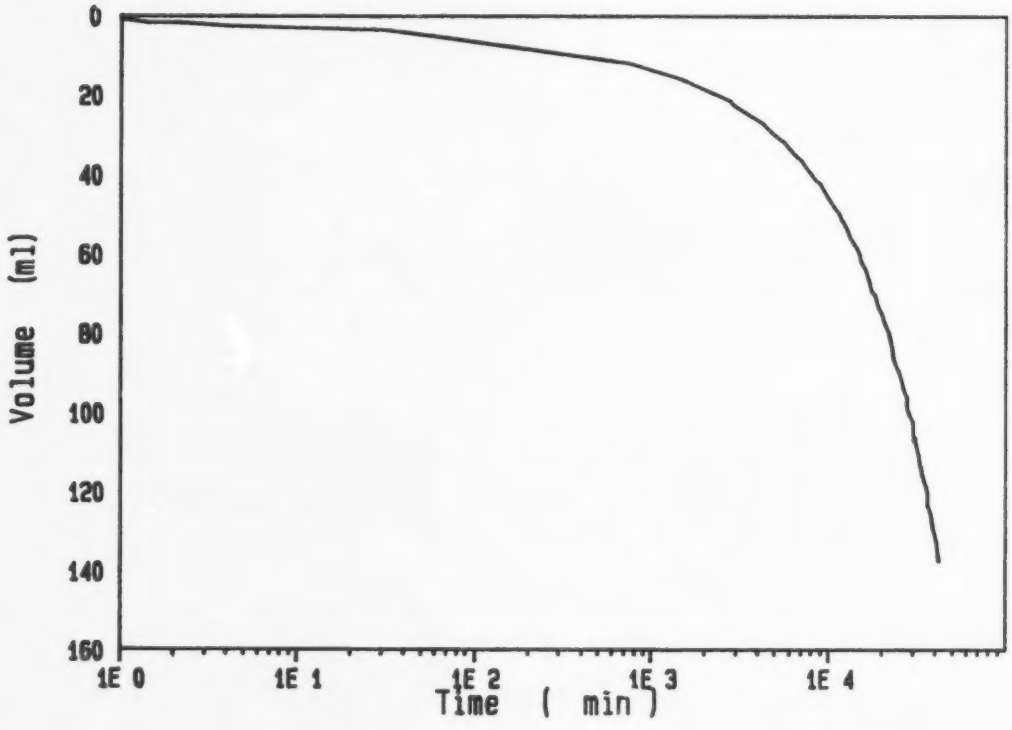

Figure C.2 Average consolidation for three bentonite plugs consolidated at 69 kPa (10 psi).

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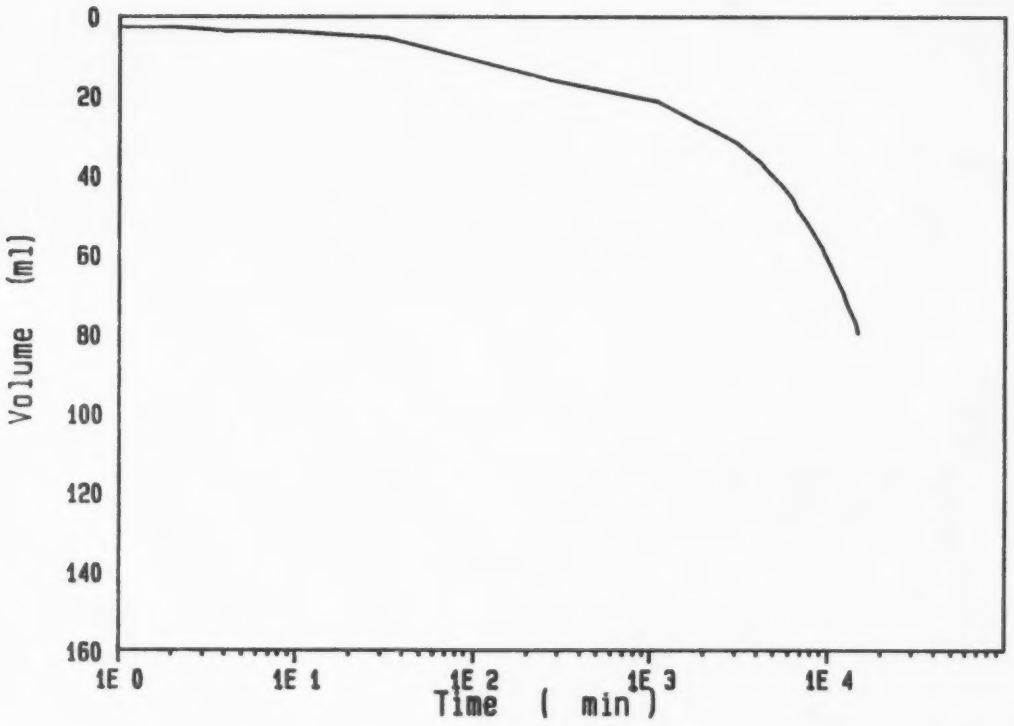

Figure C.3 Average consolidation for three bentonite plugs consolidated at 138 kPa (20 psi).

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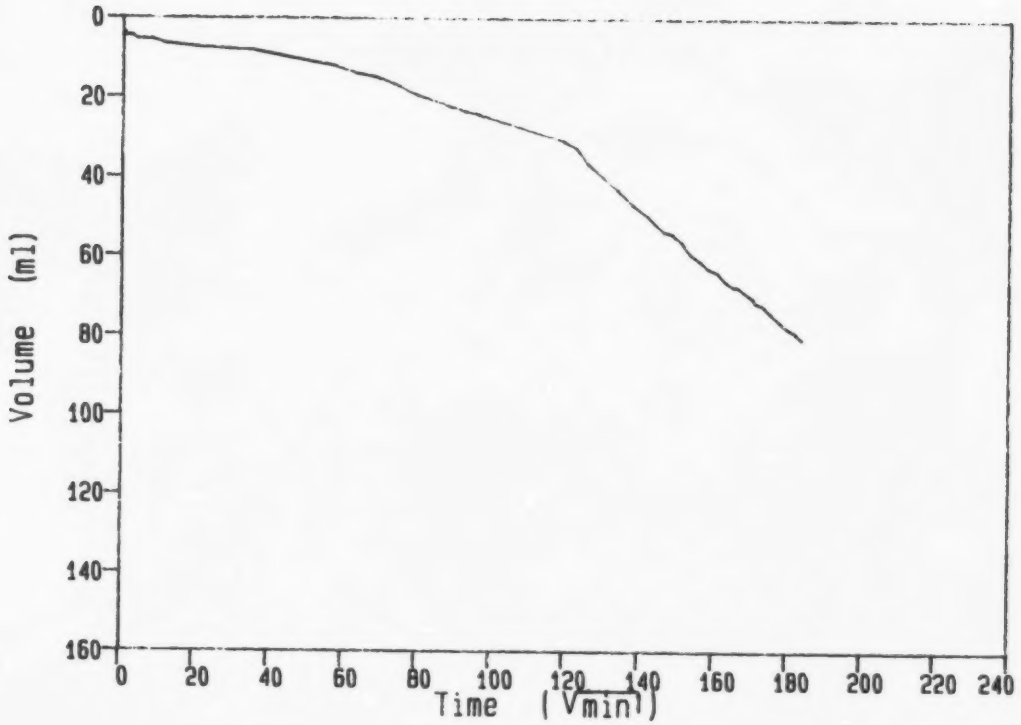

Figure C.4 Consolidation plot of Ogallala plug 1 at 34 kPa (5 psi).

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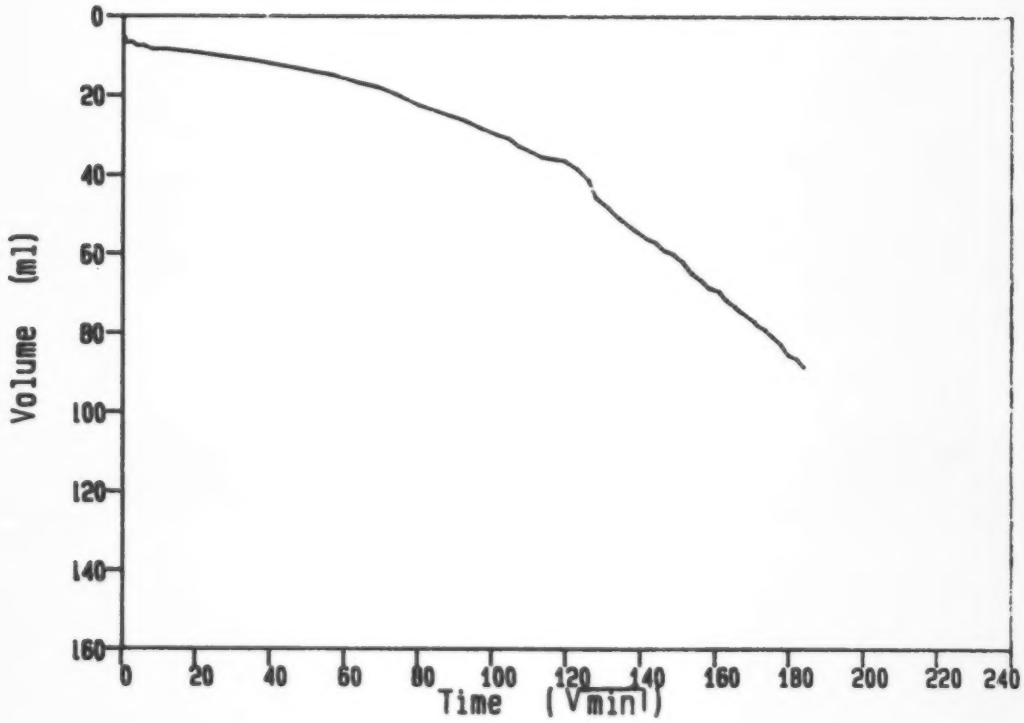

Figure C.5 Consolidation plot of Ogallala plug 2 at 34 kPa (5 psi).

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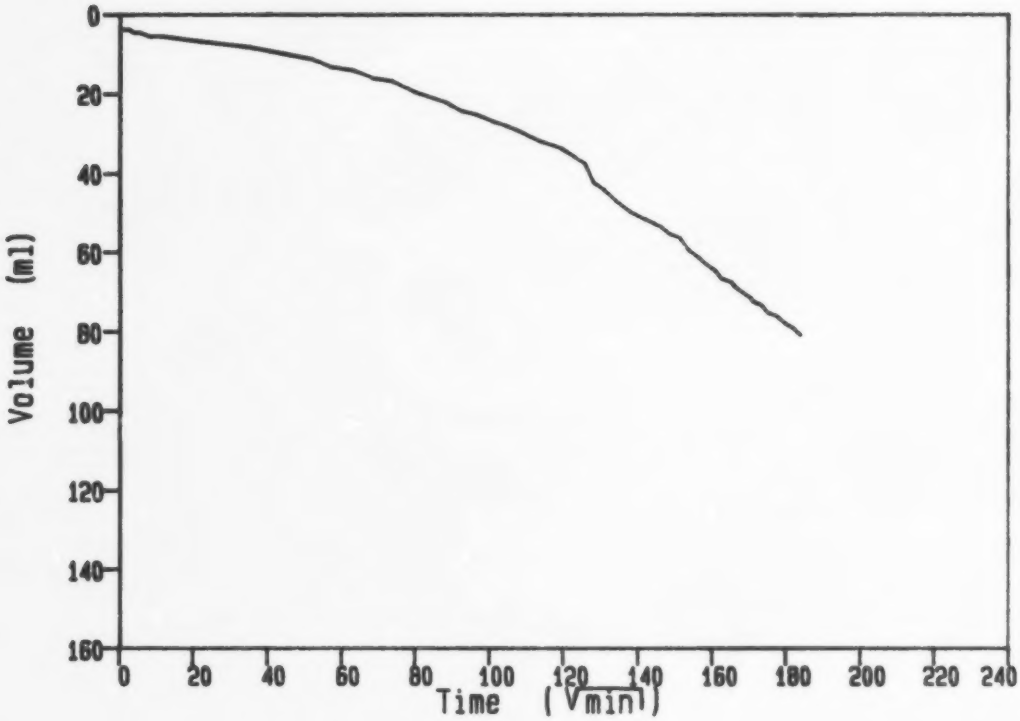

Figure C.6 Consolidation plot of Ogallala plug 3 at 34 kPa (5 psi).

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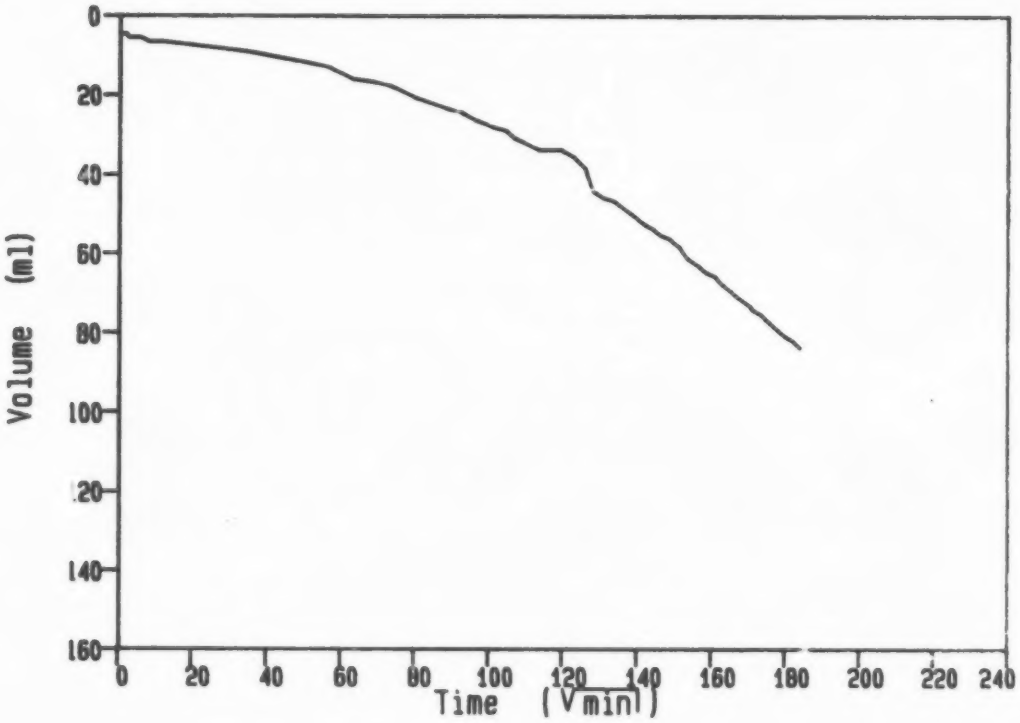

Figure C.7 Average consolidation for the three bentonite plugs consolidated at 34 kPa (5 psi).

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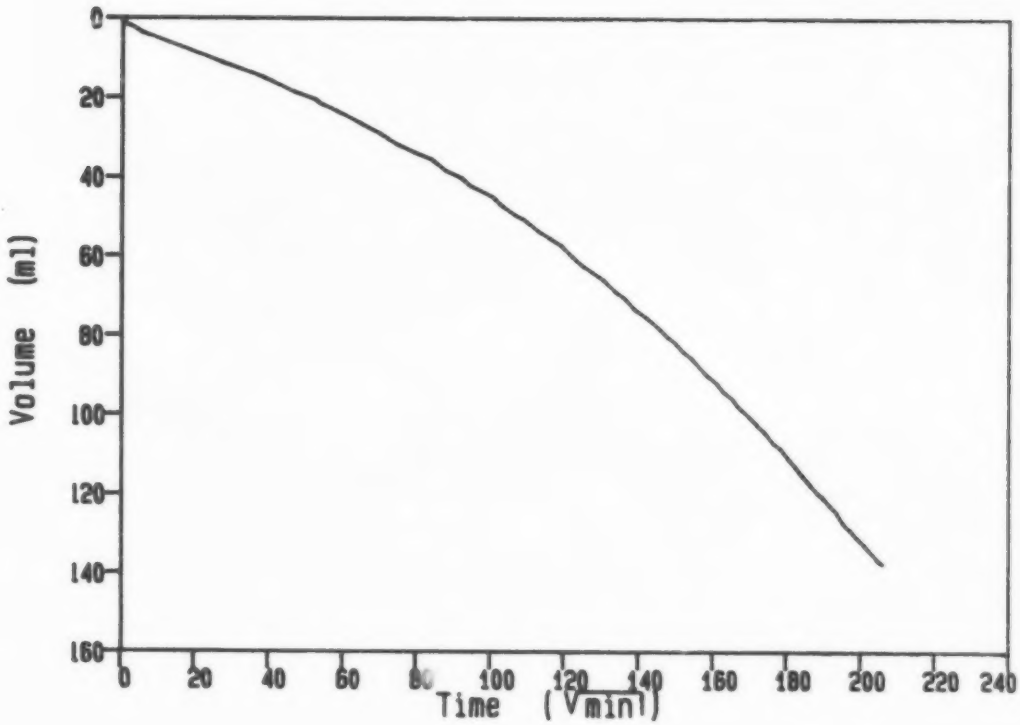

Figure C.8 Consolidation plot of Ogallala plug 1 at 69 kPa (10 psi).

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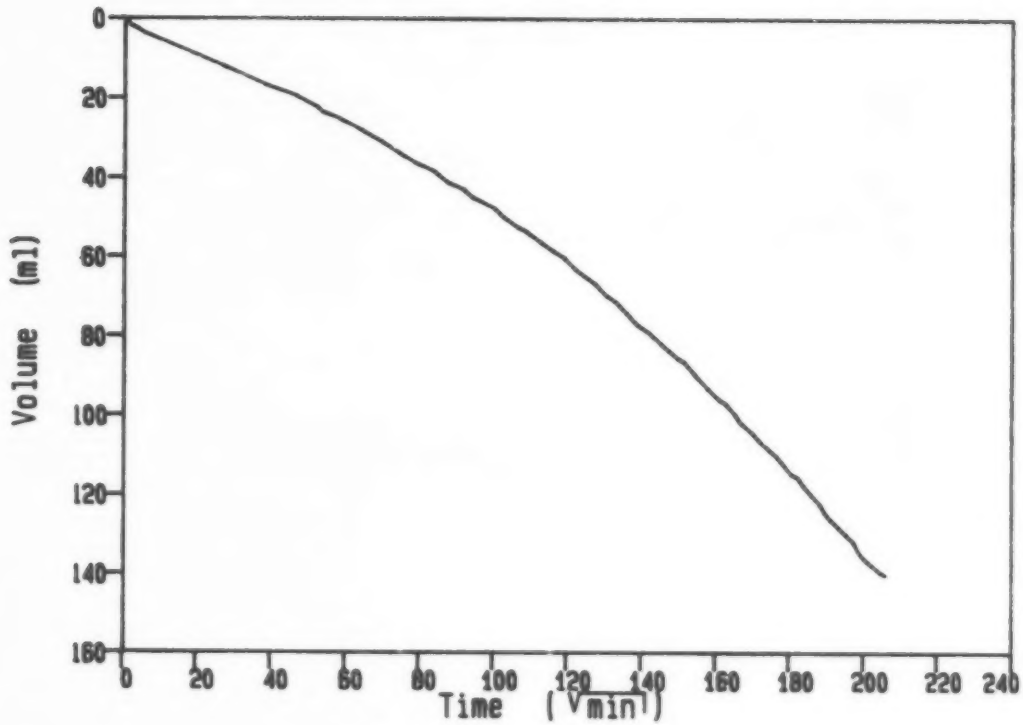

Figure C.9 Consolidation plot of Ogallala plug 2 at 69 kPa (10 psi).

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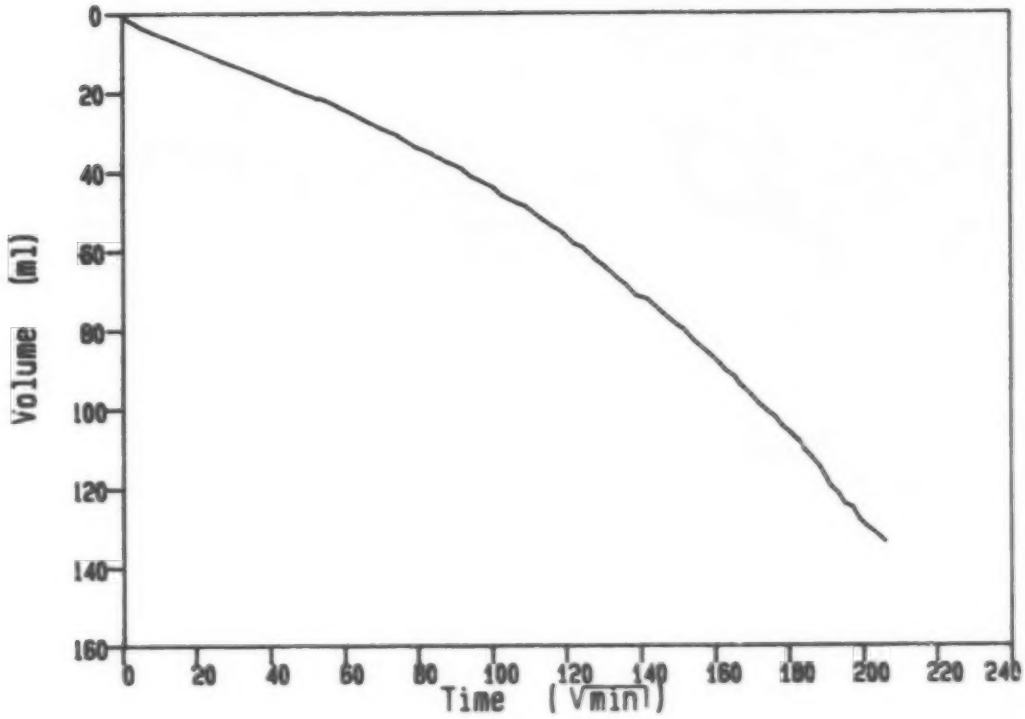

Figure C.10 Consolidation plot of Ogallala plug 3 at 69 kPa (10 psi).

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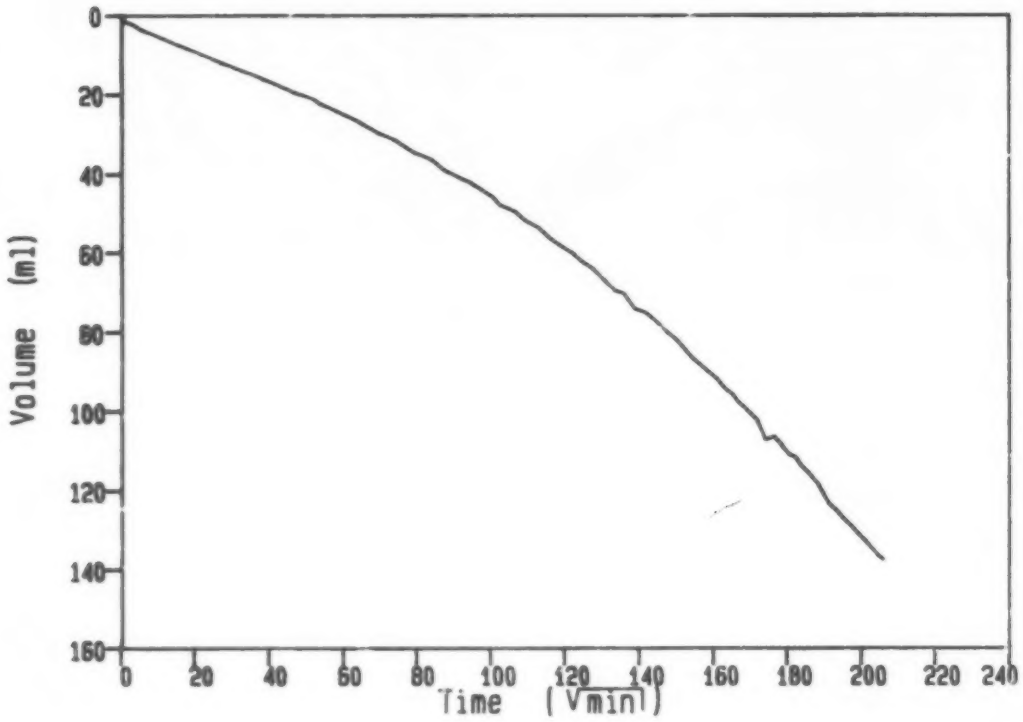

Figure C.11 Average consolidation for the three Ogallala water bentonite plugs consolidated at 69 kPa (10 psi).

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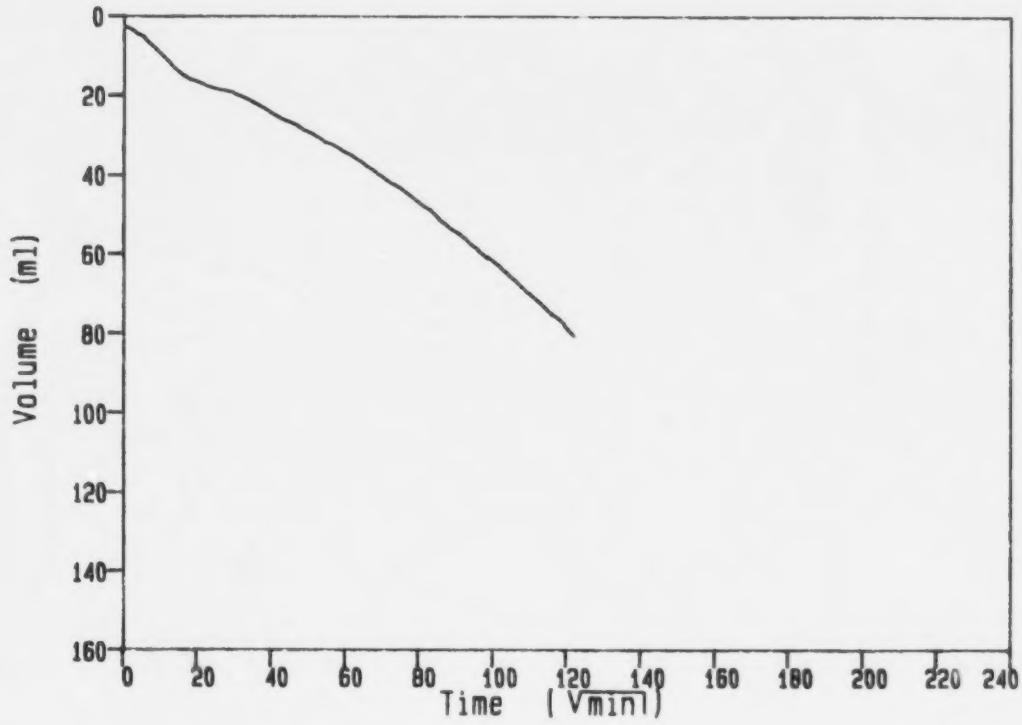

Figure C.12 Consolidation plot of Ogallala plug 1 at 138 kPa (20 psi).

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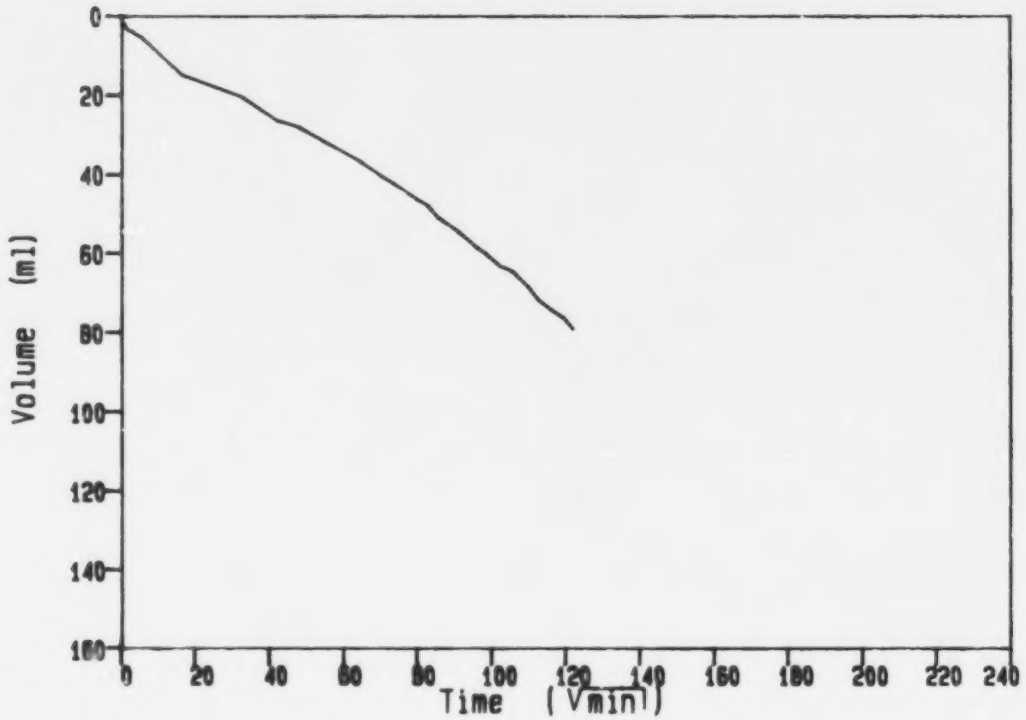

Figure C.13 Consolidation plot of Ogallala plug 2 at 138 kPa (20 psi).

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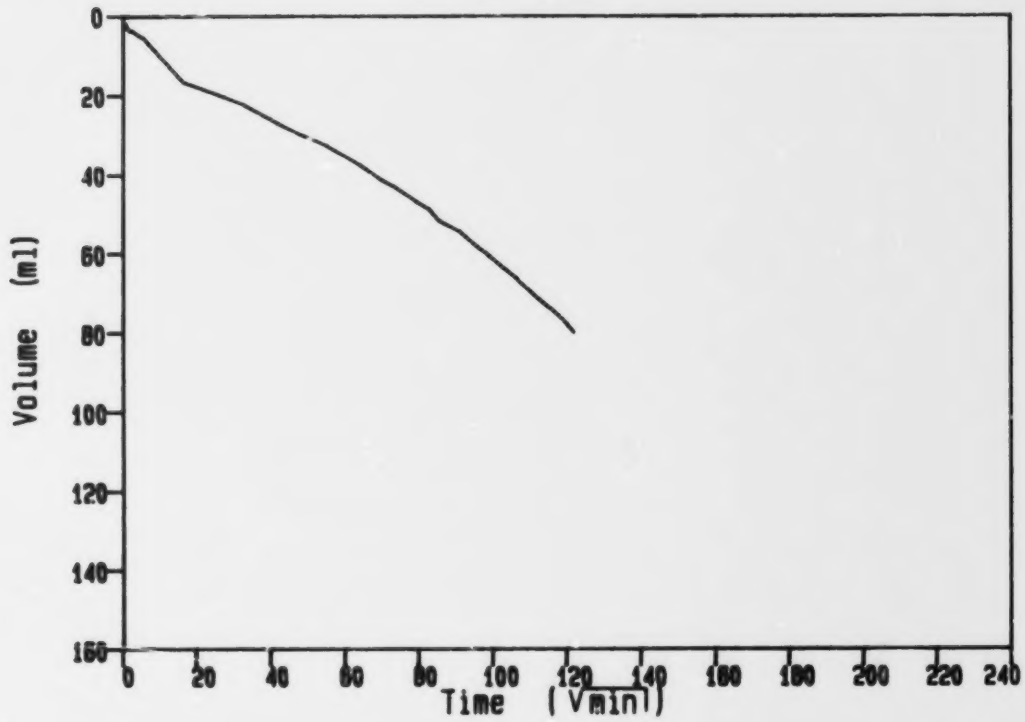

Figure C.14 Consolidation plot of Ogallala plug 3 at 138 kPa (20 psi).

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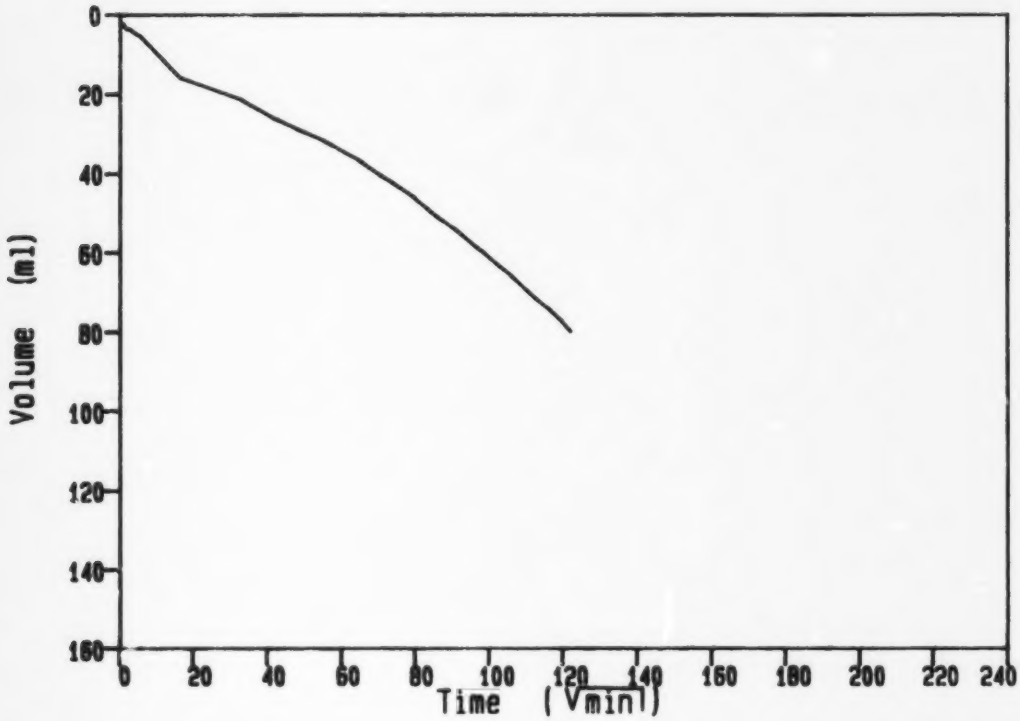

Figure C.15 Average consolidation for the three Ogallala water bentonite plugs consolidated at 138 kPa (20 psi).

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APPENDIX D

THEORETICAL TYPE CURVES AND EXPERIMENTAL HYDRAULIC HEAD DECAY CURVES
OBTAINED FROM TRANSIENT PULSE TESTING
OF BENTONITE PLUGS

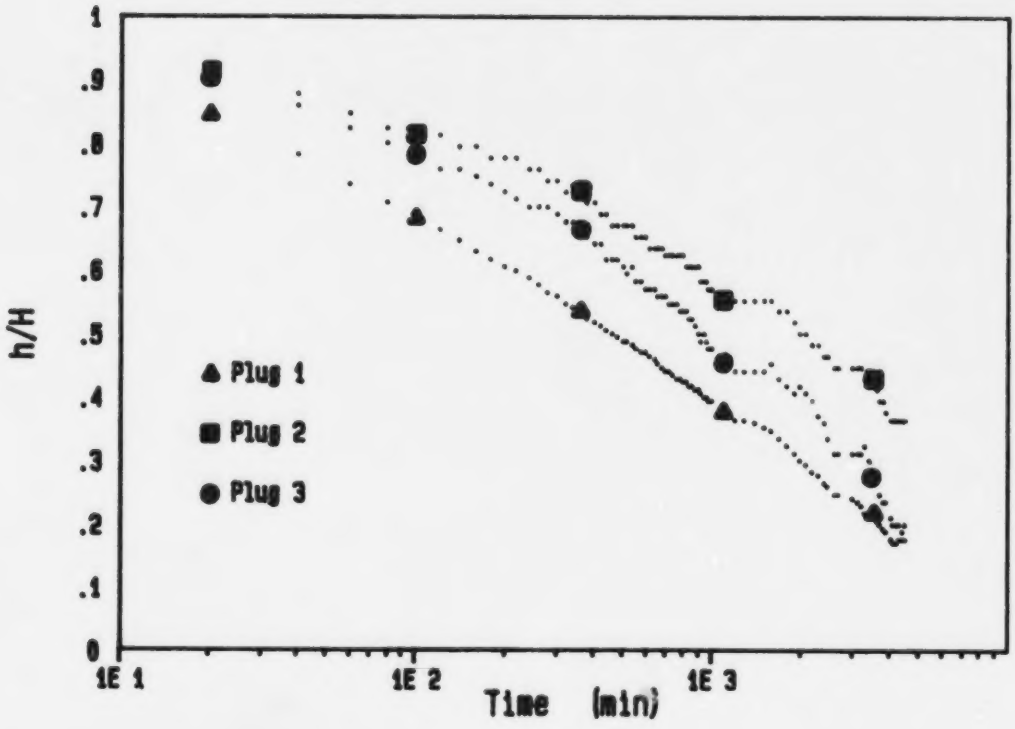

Figure D.1 Results (Decay) of the first transient pulse test conducted on the distilled water system.

H: induced head (pulse)
h: measured head

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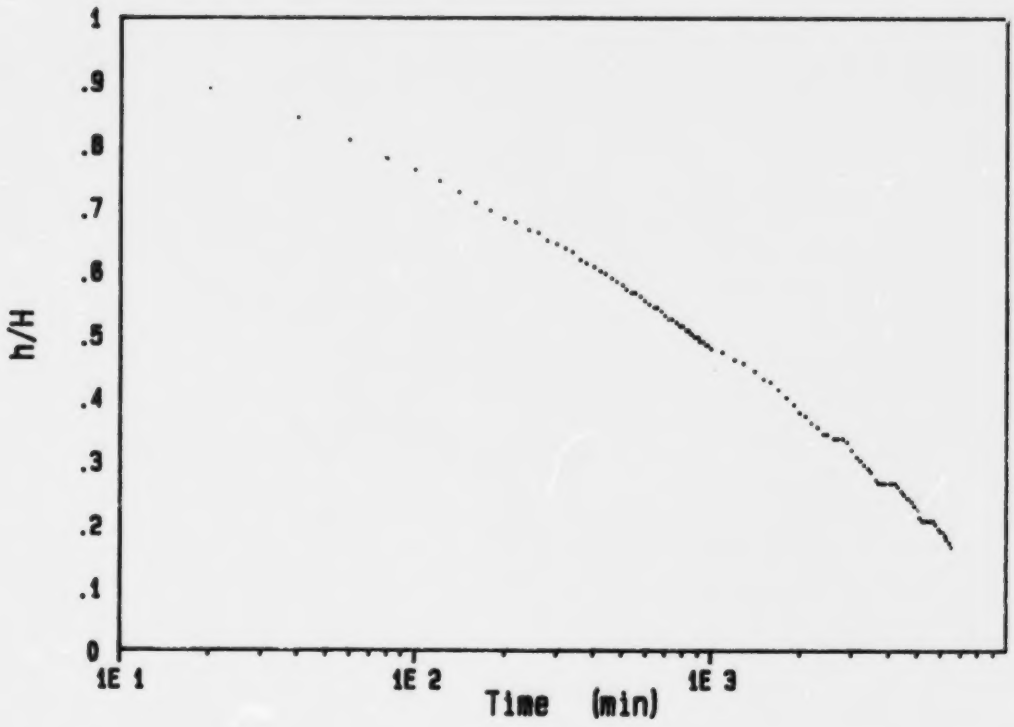

Figure D.2 Results (Decay) of the second transient pulse test conducted on plug 1 of the distilled water system.

H: induced head (pulse)
h: measured head

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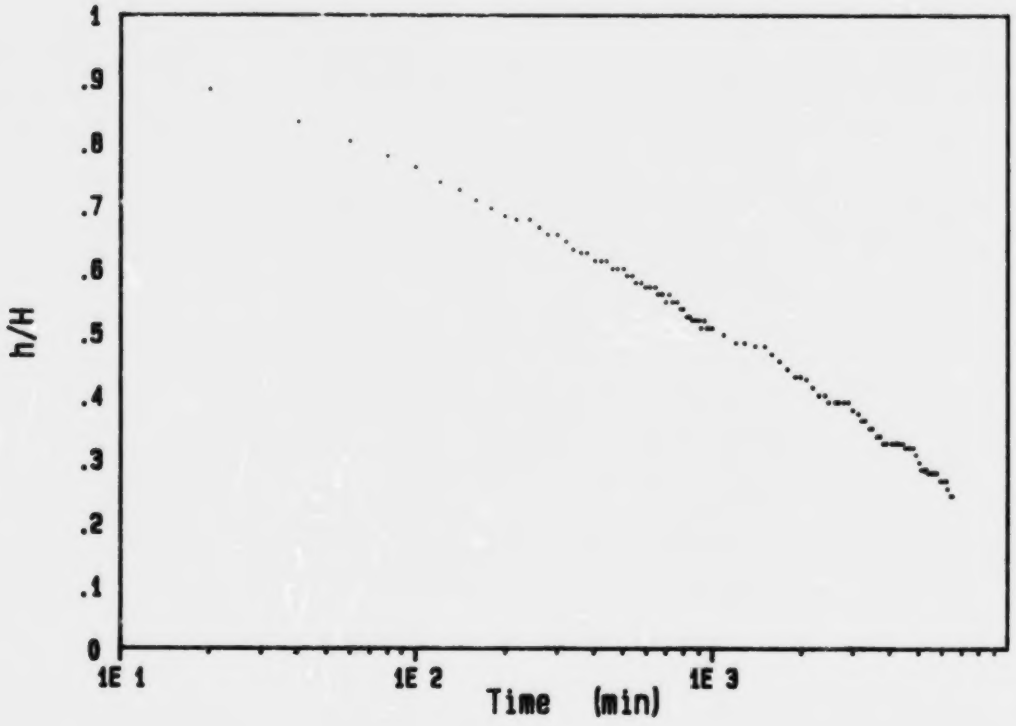

Figure D.3 Results (Decay) of the second transient pulse test conducted on plug 2 of the distilled water system.

H: induced head (pulse)
h: measured head

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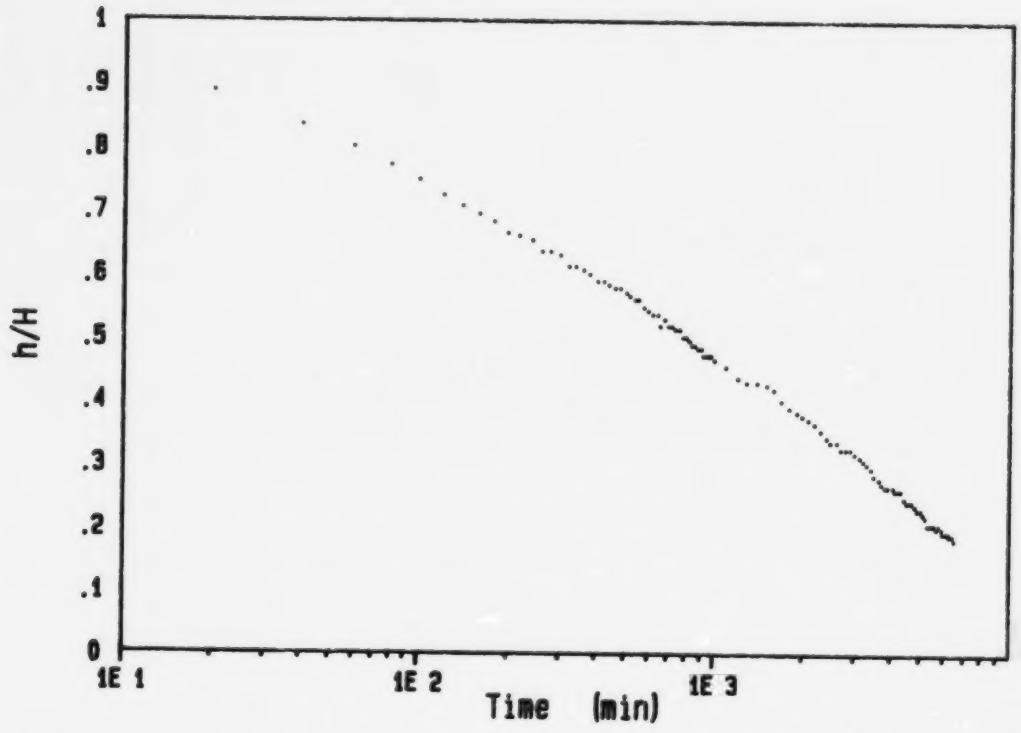

Figure D.4 Results (Decay) of the second transient pulse test conducted on plug 3 of the distilled water system.

H: induced head (pulse)
h: measured head

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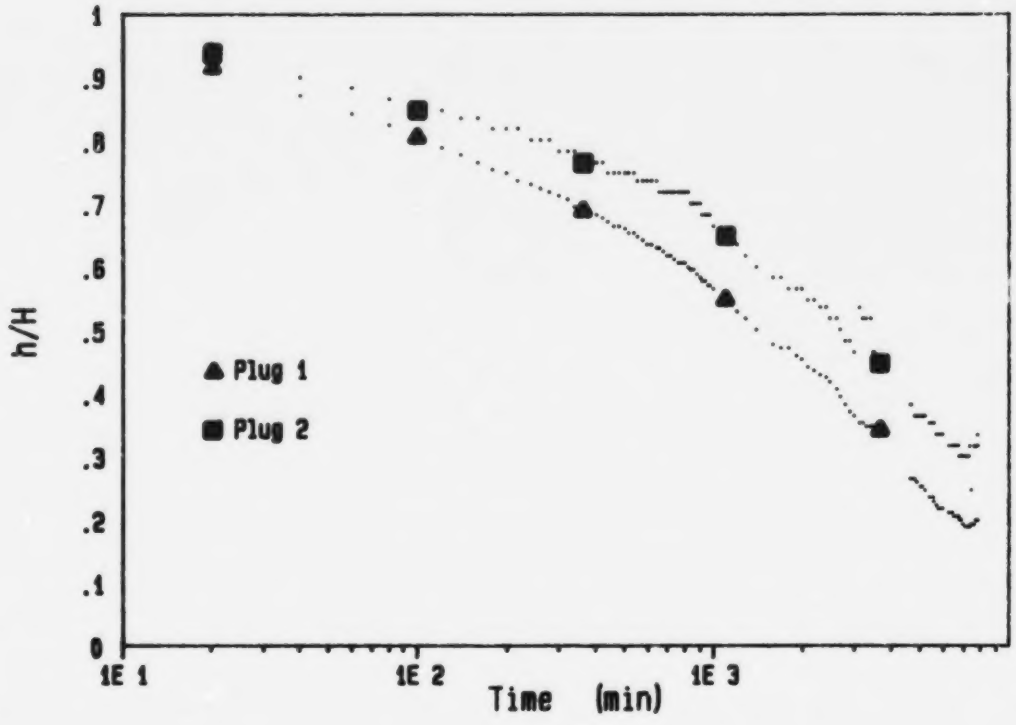

Figure D.5 Results (Decay) of the third transient pulse test conducted on plugs 1 and 2 of the distilled water system.

H: induced head (pulse)
h: measured head

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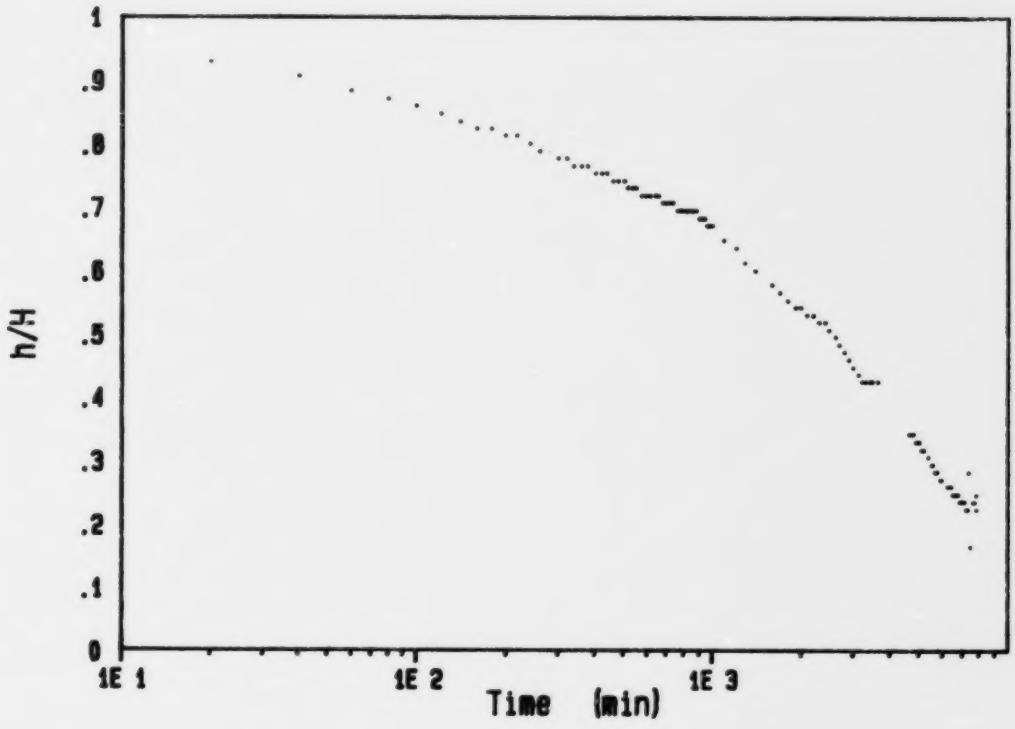

Figure D.6 Results (Decay) of the third transient pulse test conducted on plug 3 of the distilled water system.

H: induced head (pulse)
h: measured head

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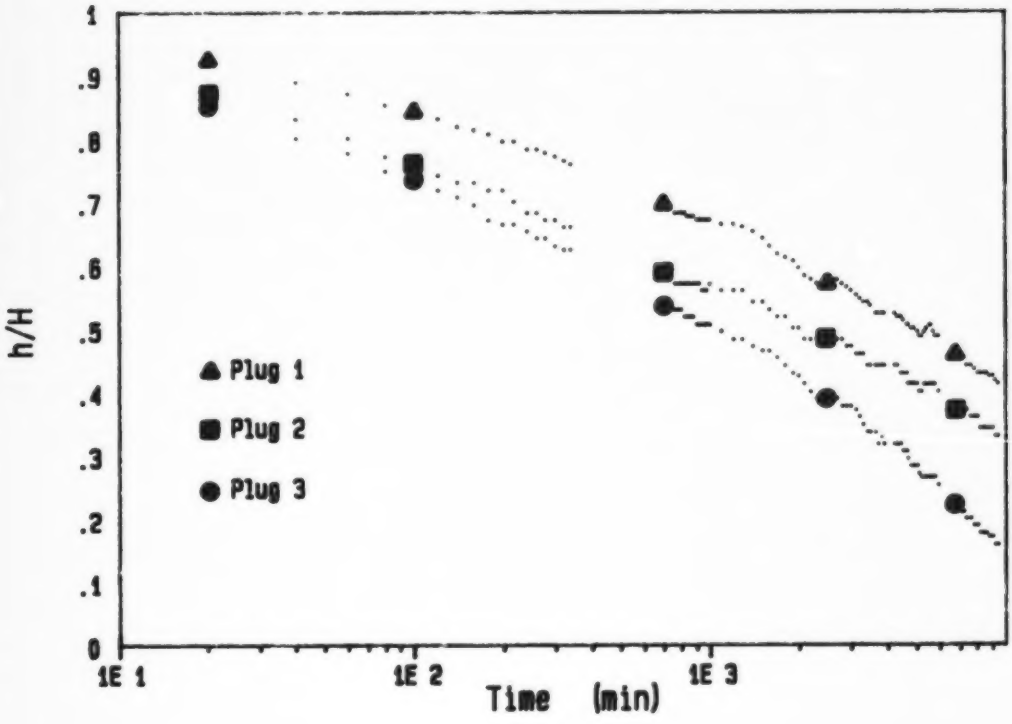

Figure D.7 Results (Decay) of the first transient pulse test conducted on the Ogallala groundwater system.

H: induced head (pulse)
h: measured head

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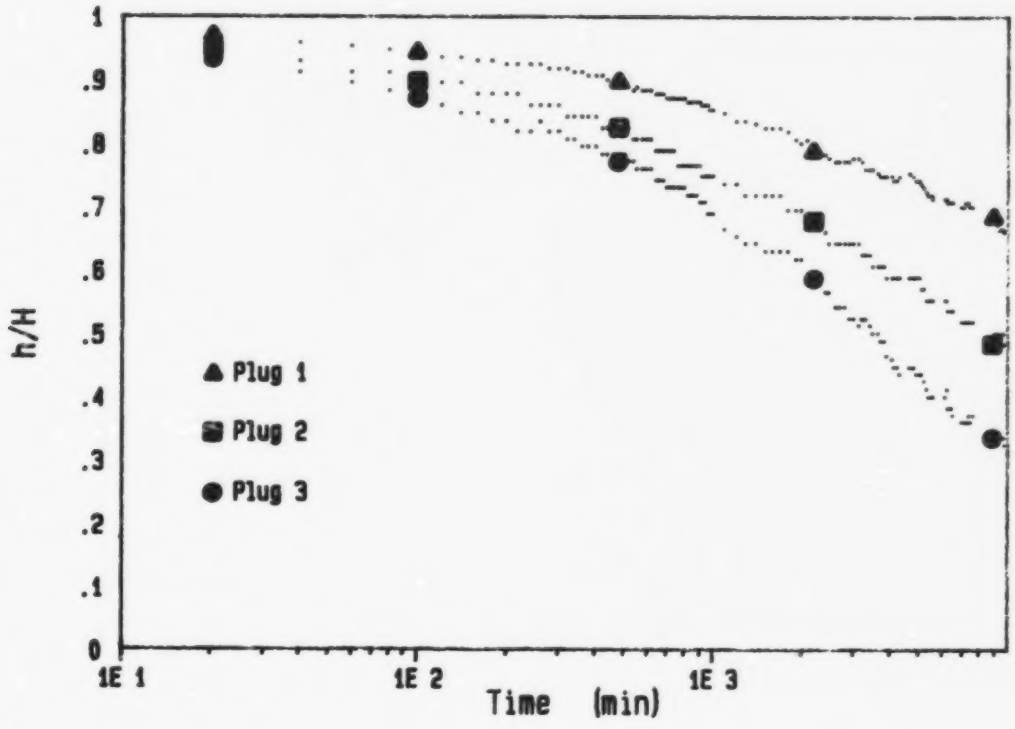

Figure D.8 Results (Decay) of the second transient pulse test conducted on the Ogallala groundwater system.

H: induced head (pulse)
h: measured head

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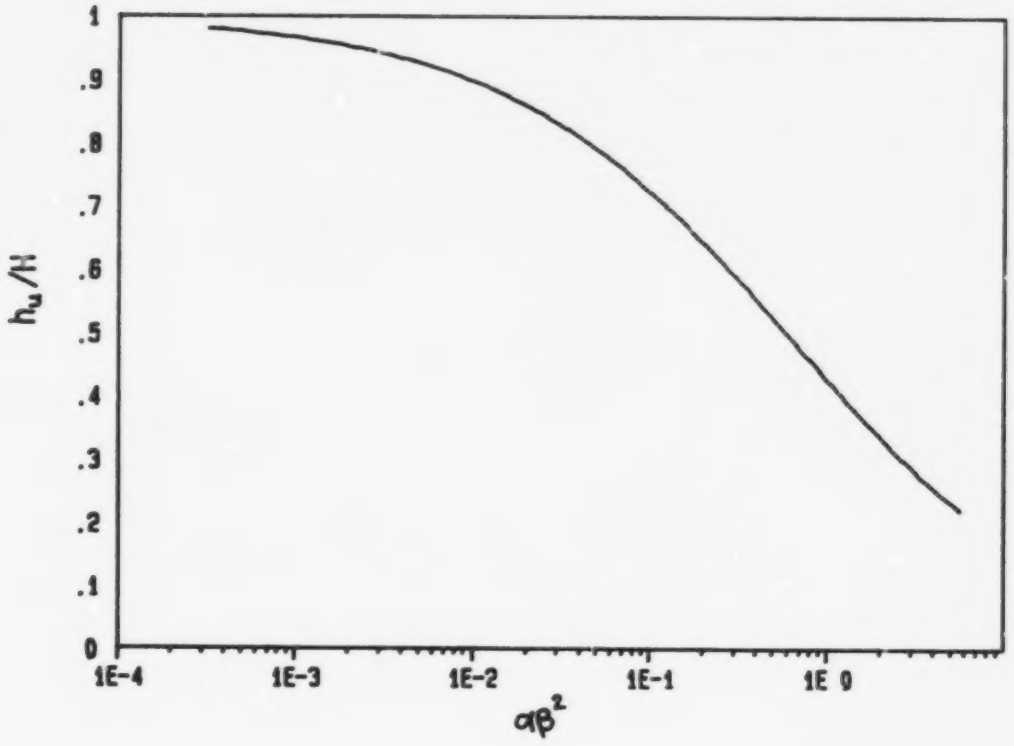

Figure D.9 Type curve for transient pulse test early time approximation.

H: induced head (pulse)
h_u: monitored upstream reservoir head
α: dimensionless time
β: reservoir compressive storage ratio

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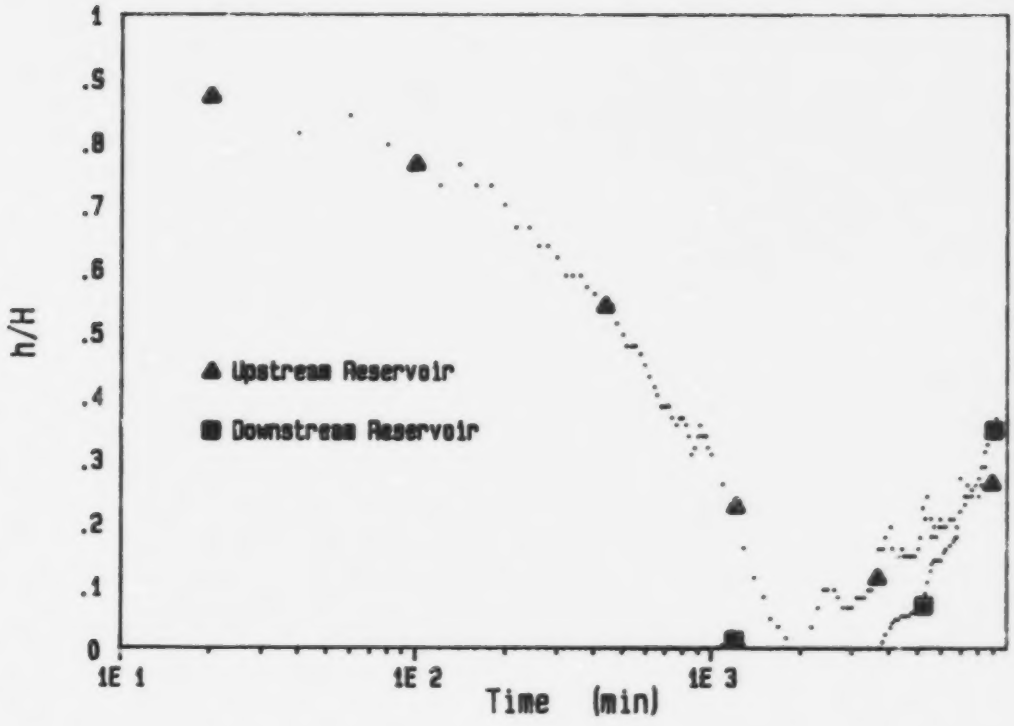

Figure D.10 Hydraulic head decay and build up curves resulting from injection into the small upstream reservoir of the distilled water system consisting of a 1 cm long bentonite plug having an initial moisture content of 75 percent.

H: induced head (pulse)
h: monitored head

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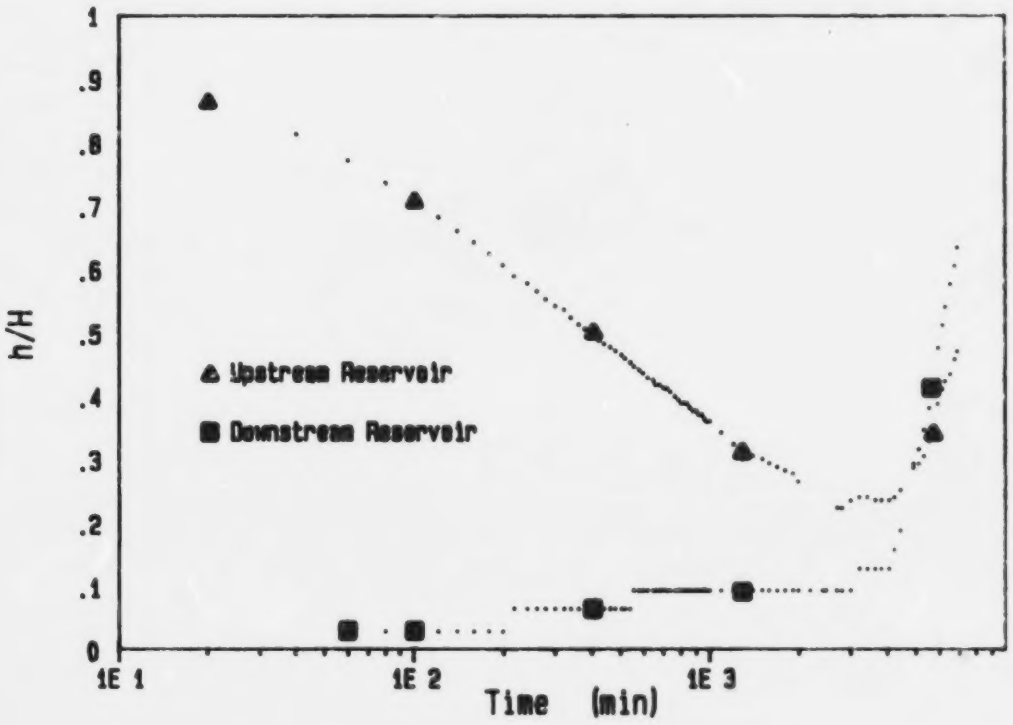

Figure D.11 Hydraulic head decay and build up curves resulting from injection into the small upstream reservoir of the distilled water system consisting of a 1 cm long bentonite plug having an initial moisture content of 125 percent.

H: induced head (pulse)
h: monitored head

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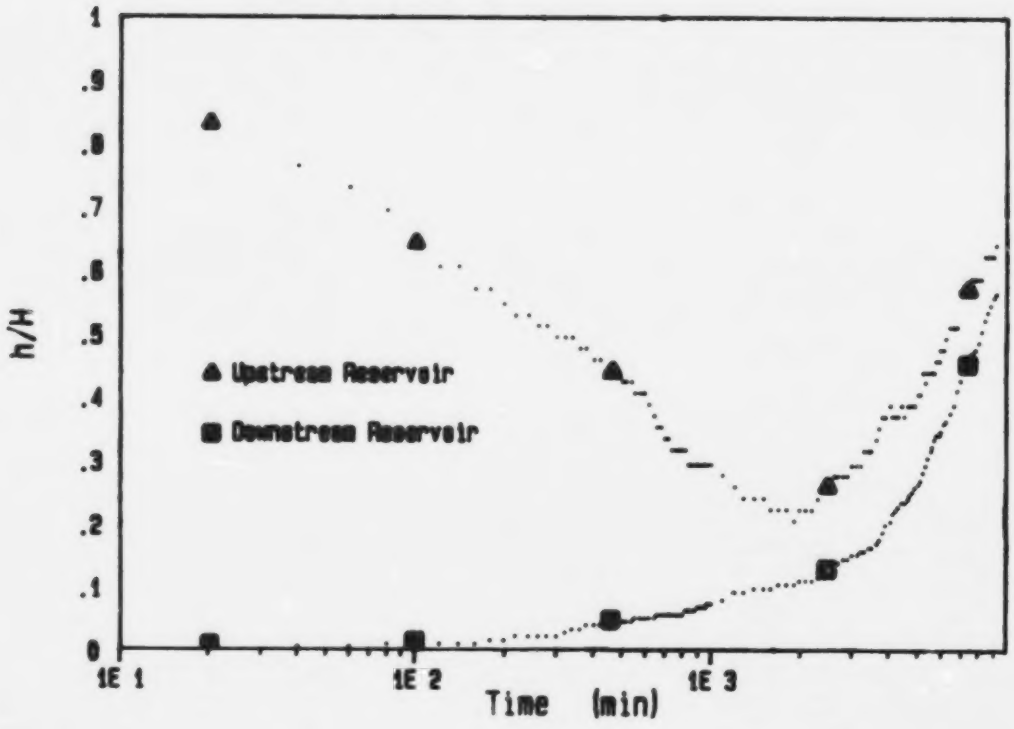

Figure D.12 Hydraulic head decay and build up curves resulting from injection into the small upstream reservoir of the Ogallala groundwater system consisting of a 1 cm long bentonite plug having an initial moisture content of 75 percent.

H: induced head (pulse)
h: monitored head

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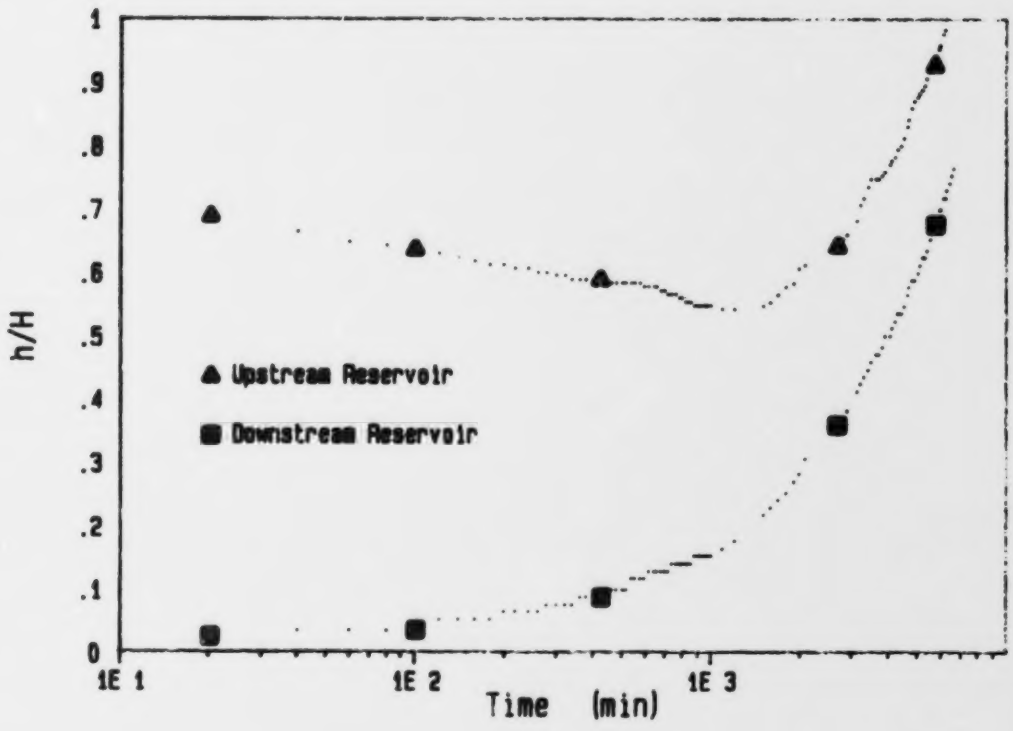

Figure D.13 Hydraulic head decay and build up curves resulting from injection into the small upstream reservoir of the Ogallala groundwater system consisting of a 1 cm long bentonite plug having an initial moisture content of 100 percent.

H: induced head (pulse)
h: monitored head

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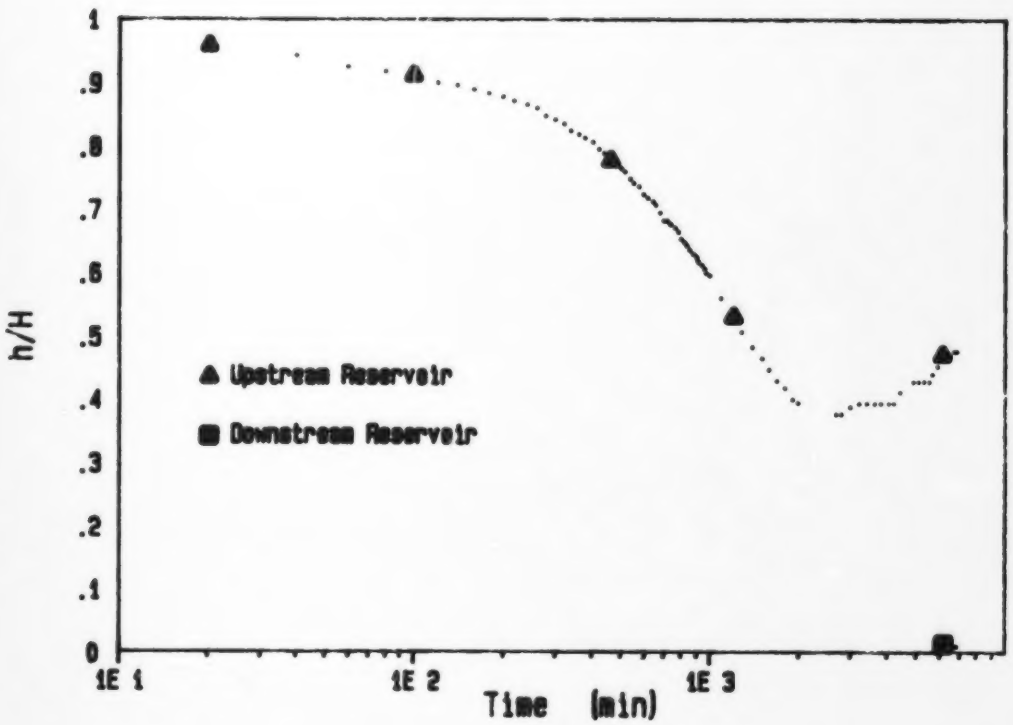

Figure D.14 Hydraulic head decay and build up curves resulting from injection into the small upstream reservoir of the Ogallala groundwater system consisting of a 1 cm long bentonite plug having an initial moisture content of 125 percent.

H: induced head (pulse)
h: monitored head

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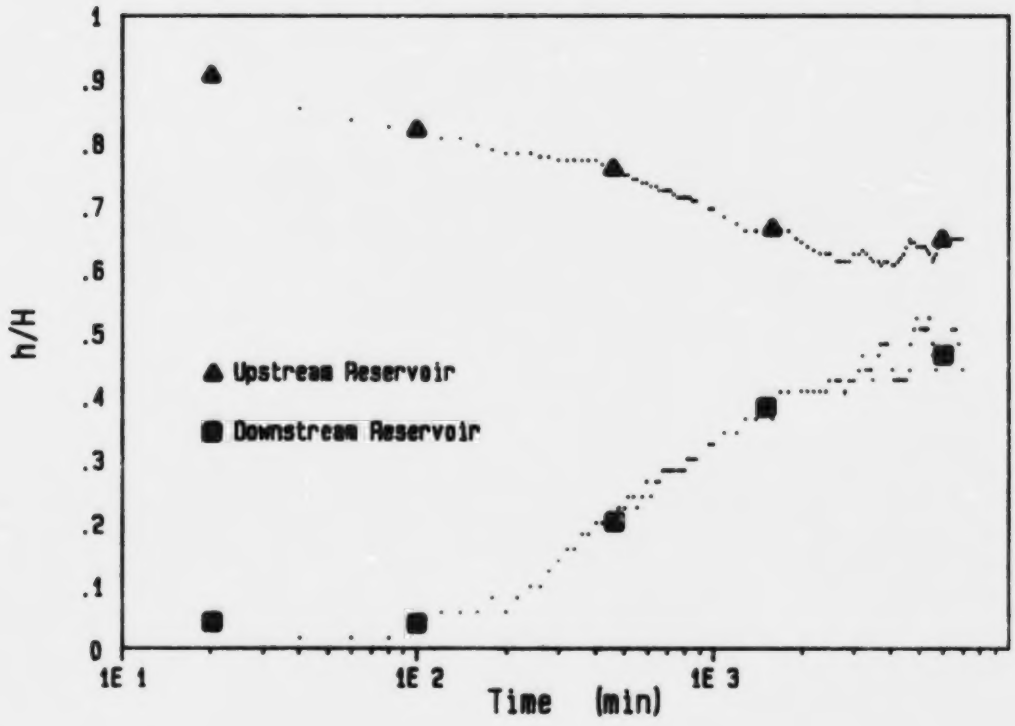

Figure D.15 Hydraulic head decay and build up curves resulting from injection into the large upstream reservoir of the distilled water system consisting of a 1 cm long bentonite plug having an initial moisture content of 75 percent.

H: induced head (pulse)
h: monitored head

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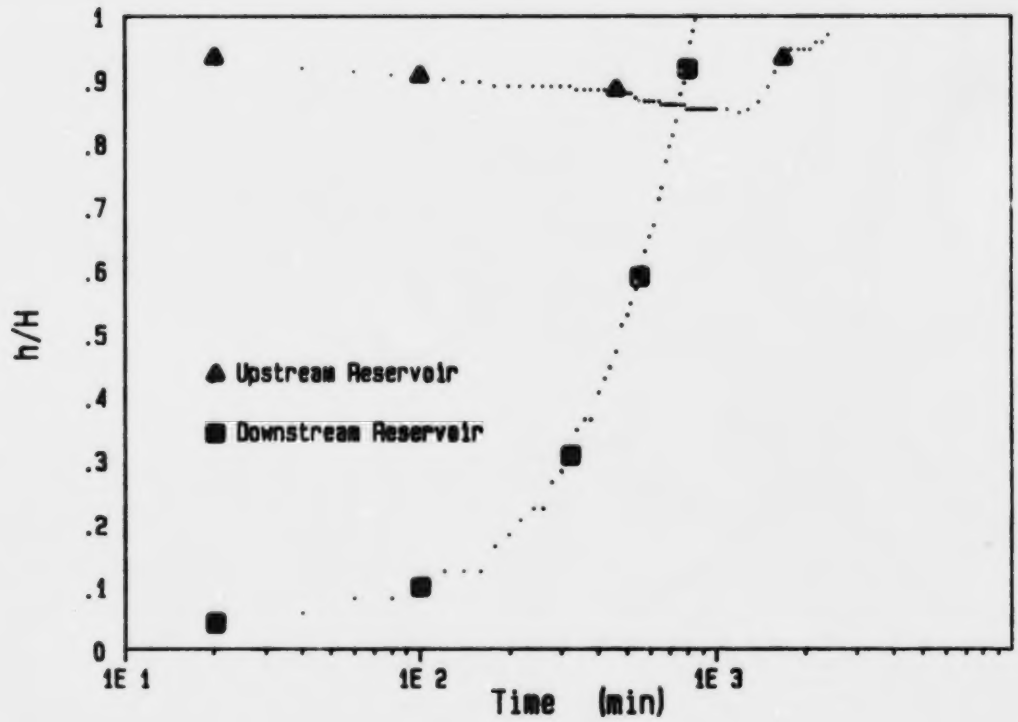

Figure D.16 Hydraulic head decay and build up curves resulting from injection into the large upstream reservoir of the distilled water system consisting of a 1 cm long bentonite plug having an initial moisture content of 125 percent.

H: induced head (pulse)
h: monitored head

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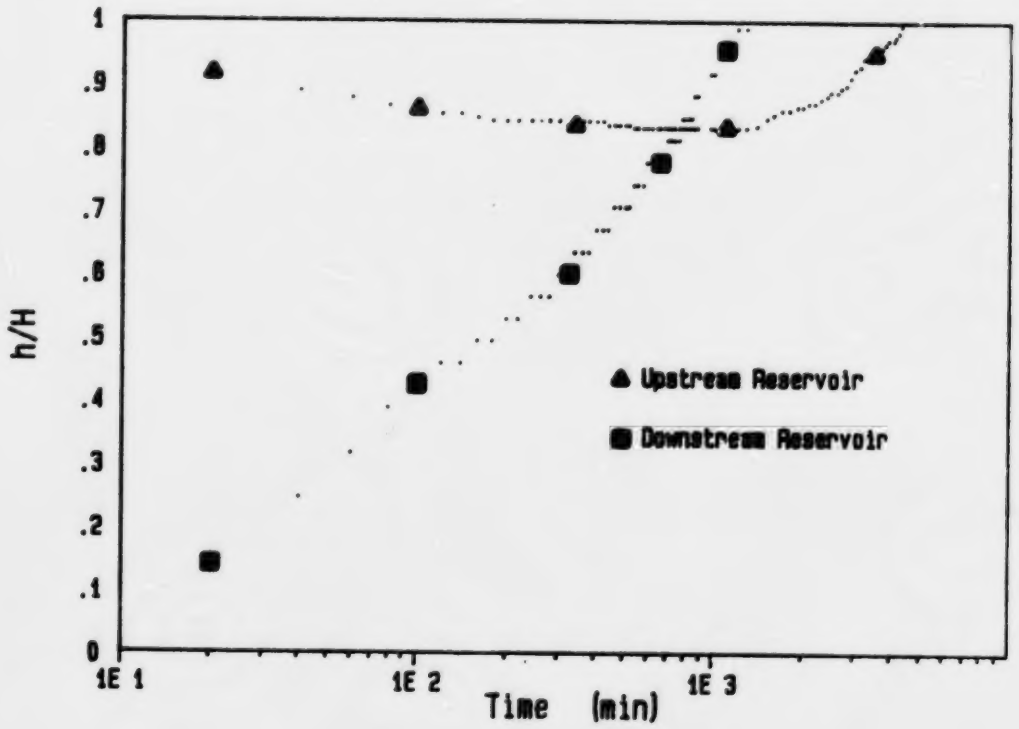

Figure D.17 Hydraulic head decay and build up curves resulting from injection into the large upstream reservoir of the Ogallala groundwater system consisting of a 1 cm long bentonite plug having an initial moisture content of 75 percent.

H: induced head (pulse)
h: monitored head

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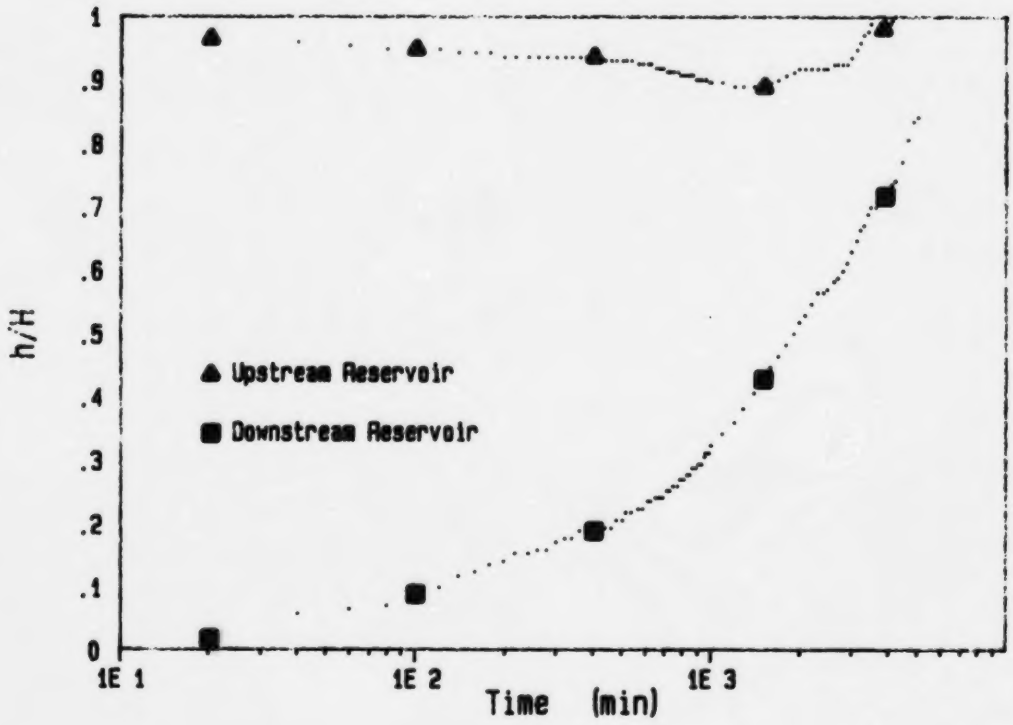

Figure D.18 Hydraulic head decay and build up curves resulting from injection into the large upstream reservoir of the Ogallala groundwater system consisting of a 1 cm long bentonite plug having an initial moisture content of 100 percent.

H: induced head (pulse)
h: monitored head

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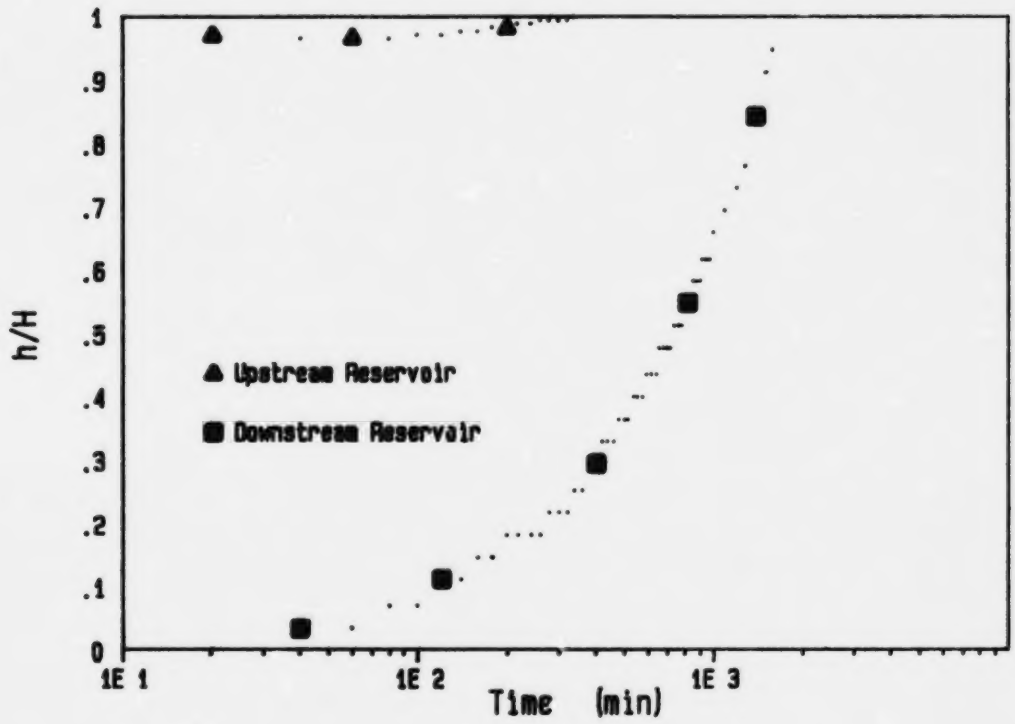

Figure D.19 Hydraulic head decay and build up curves resulting from injection into the large upstream reservoir of the Ogallala groundwater system consisting of a 1 cm long bentonite plug having an initial moisture content of 125 percent.

H: induced head (pulse)
h: monitored head

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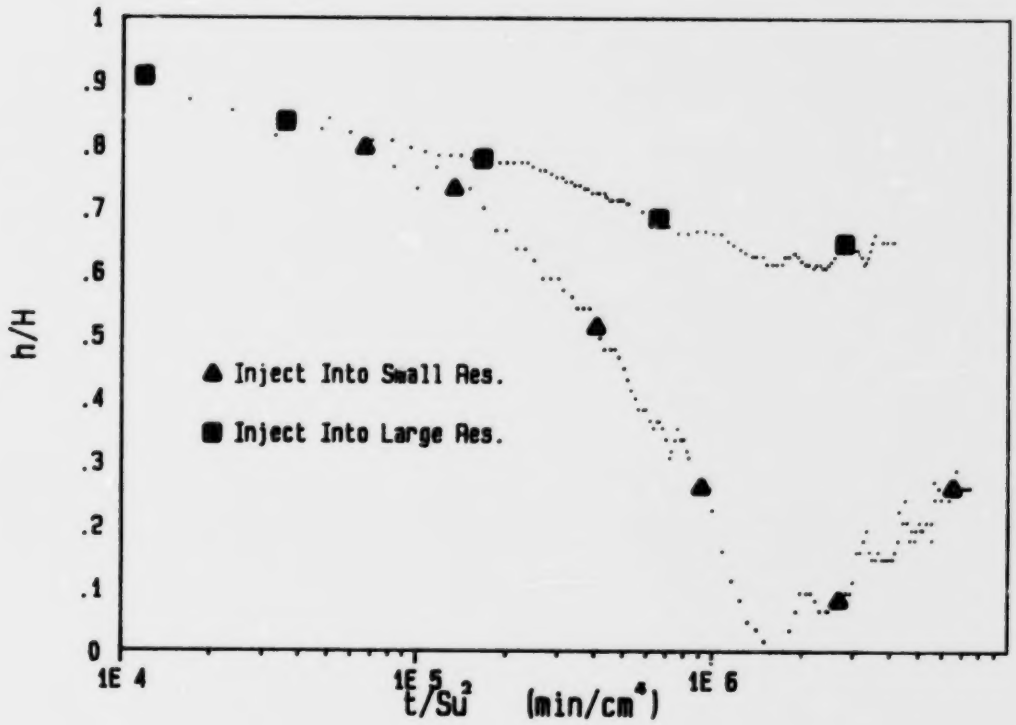

Figure D.20 Hydraulic head decay as a function of the compressive storage of the upstream reservoir of the distilled water system consisting of a 1 cm long bentonite plug having an initial moisture content of 75 percent.

H: induced head (pulse)
h: monitored head
S_u: upstream reservoir storage

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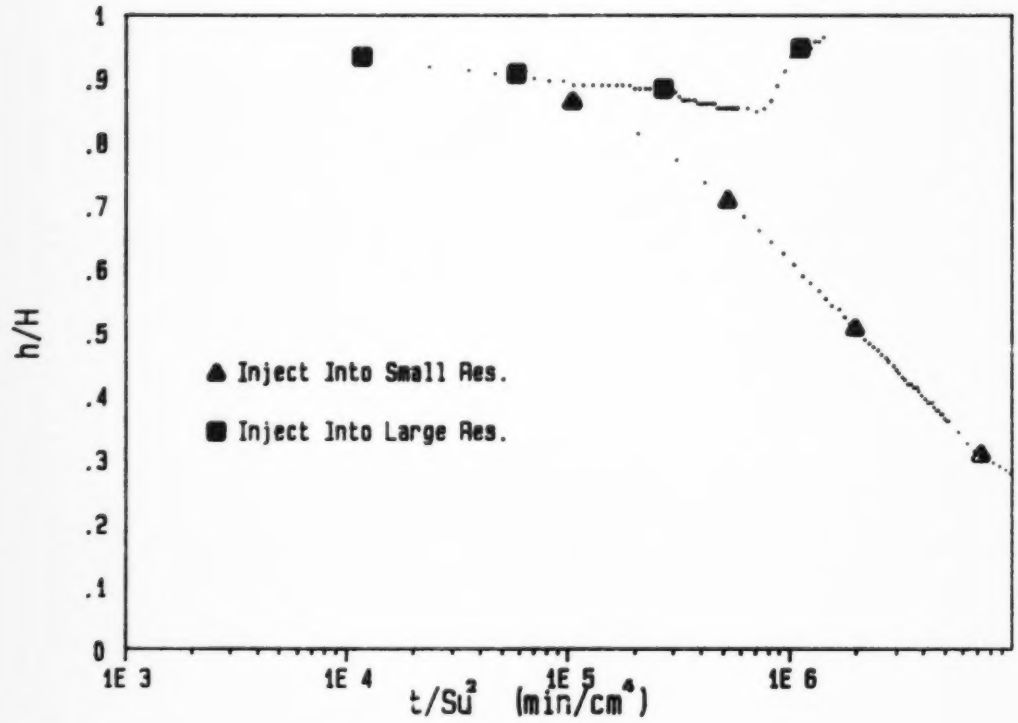

Figure D.21 Hydraulic head decay as a function of the compressive storage of the upstream reservoir of the distilled water system consisting of a 1 cm long bentonite plug having an initial moisture content of 125 percent.

H: induced head (pulse)
h: monitored head
S_u: upstream reservoir storage

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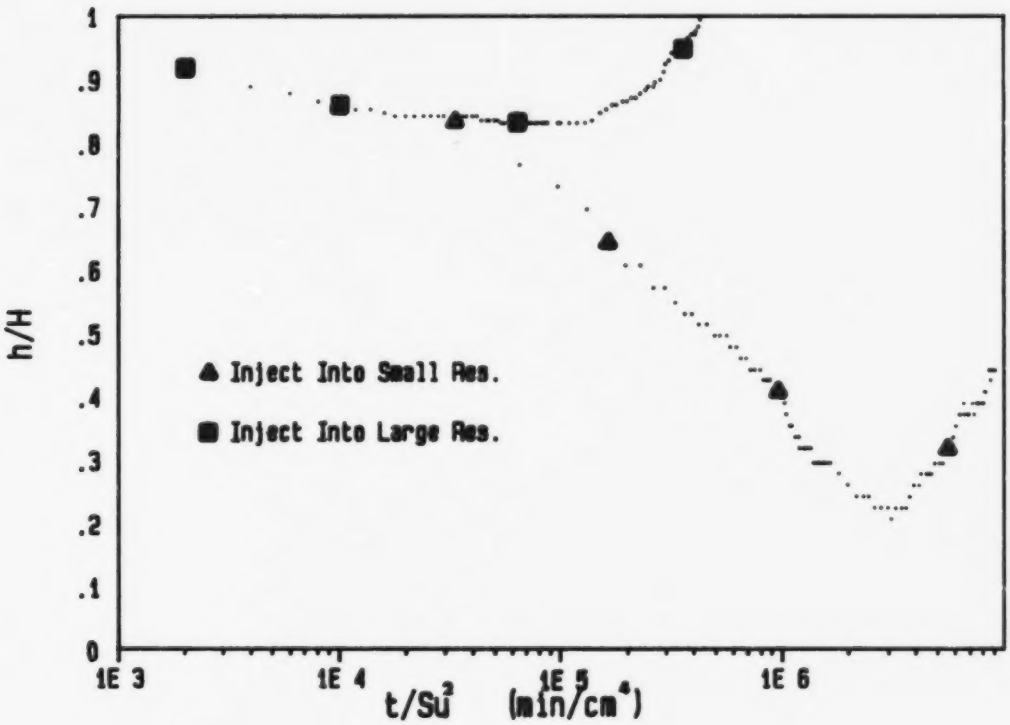

Figure D.22 Hydraulic head decay as a function of the compressive storage of the upstream reservoir of the Ogallala groundwater system consisting of a 1 cm long bentonite plug having an initial moisture content of 75 percent.

H: induced head (pulse)
h: monitored head
S_u: upstream reservoir storage

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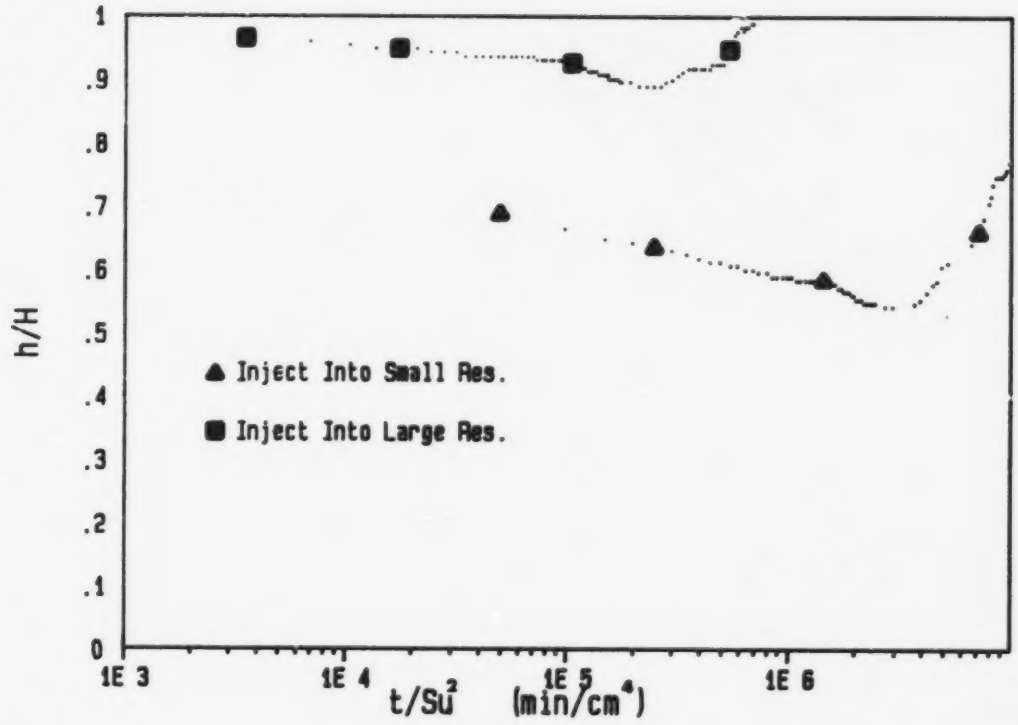

Figure D.23 Hydraulic head decay as a function of the compressive storage of the upstream reservoir of the Ogallala groundwater system consisting of a 1 cm long bentonite plug having an initial moisture content of 100 percent.

H: induced head (pulse)
h: monitored head
S_u: upstream reservoir storage

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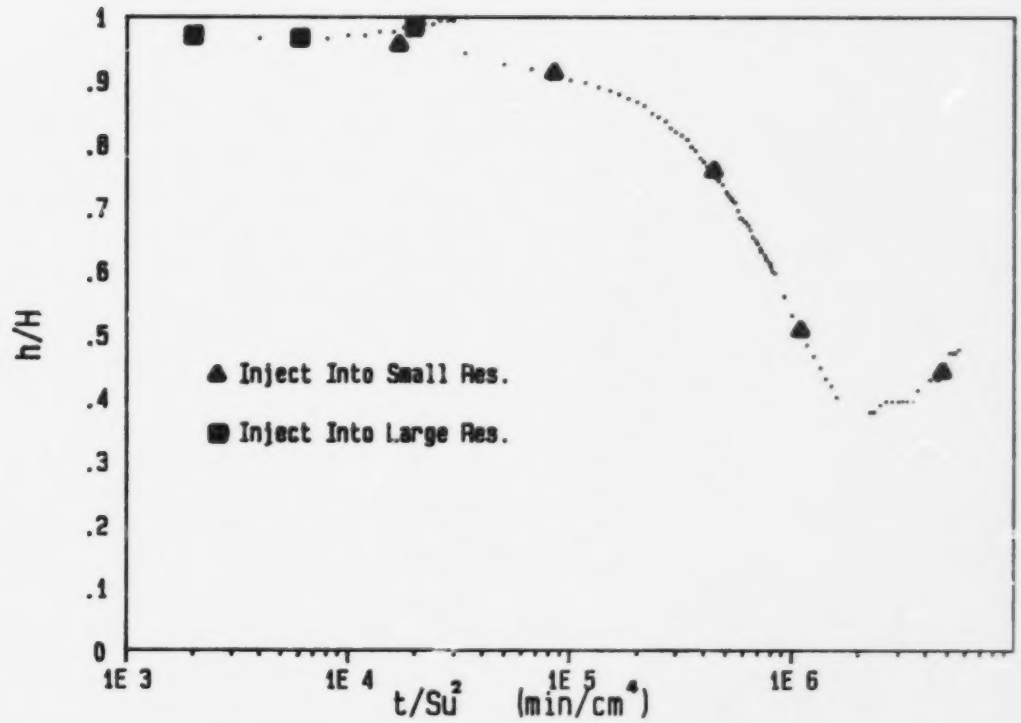

Figure D.24 Hydraulic head decay as a function of the compressive storage of the upstream reservoir of the Ogallala groundwater system consisting of a 1 cm long bentonite plug having an initial moisture content of 125 percent.

H: induced head (pulse)
h: monitored head
S_u: upstream reservoir storage

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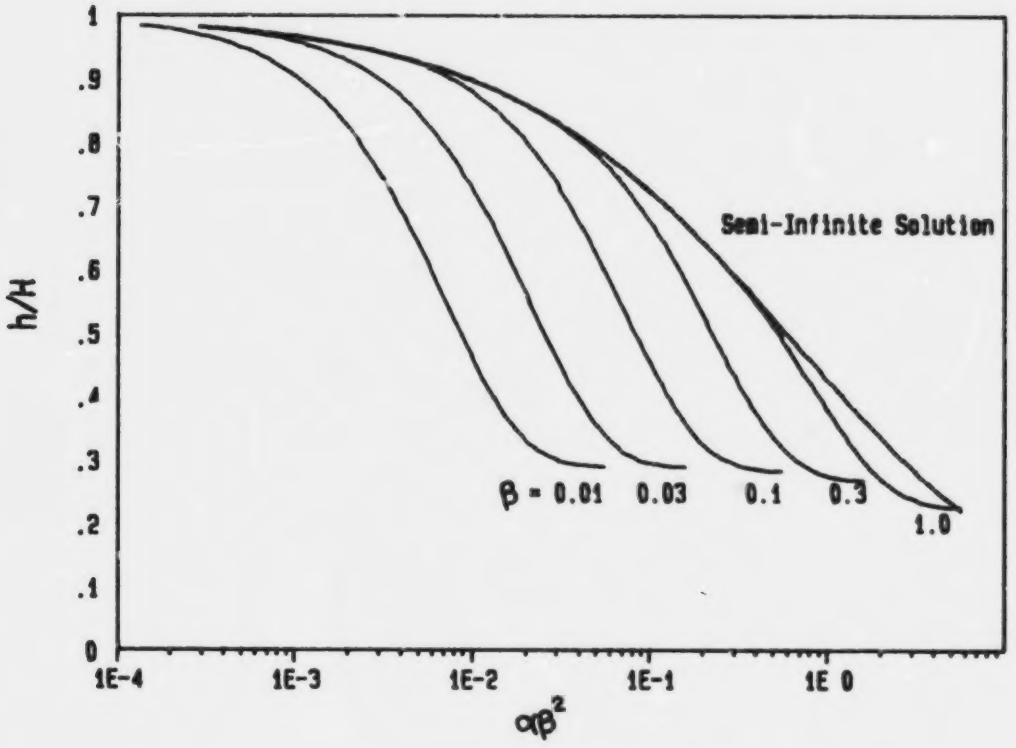

Figure D.25 Transient pulse testing type curves for $\gamma = 2.44$. Plot of h/H vs. $\alpha\beta^2$ for various values of β (Section 4.3.2).

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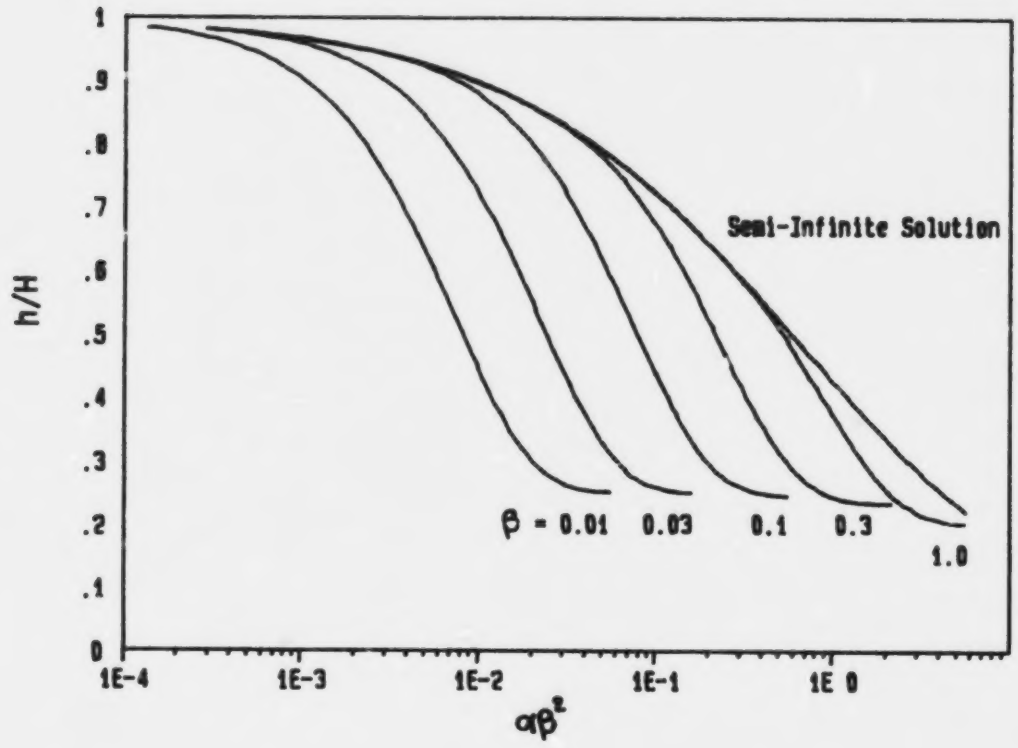

Figure D.26 Transient pulse testing type curves for $\gamma = 2.97$. Plot of h/H vs. $\alpha\beta^2$ for various values of β (Section 4.3.2).

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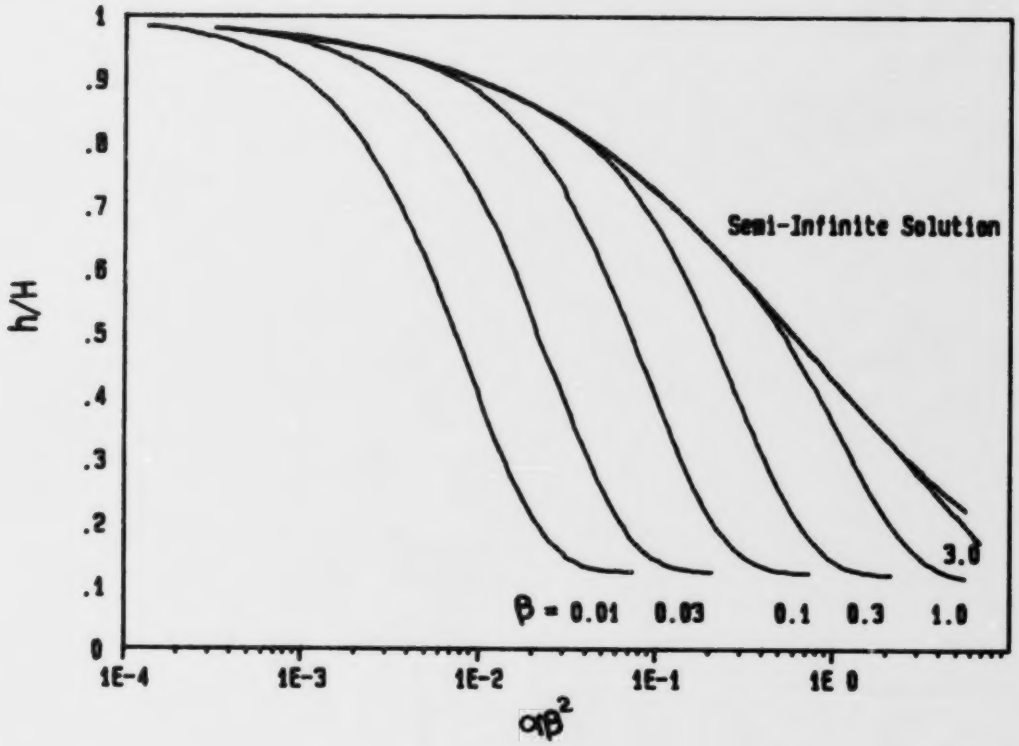

Figure D.27 Transient pulse testing type curves for $\gamma = 7.15$. Plot of h/H vs. $\alpha\beta^2$ for various values of β (Section 4.3.2).

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14 ABSTRACT (200 words or less)

The hydraulic conductivity has been determined of plugs constructed with commercial precompressed bentonite pellets. Bentonite has been hydrated and tested with waters of five different chemical compositions, including one groundwater (Ogallala aquifer, Texas). The groundwater contained a significant amount of solids; waters prepared in the laboratory did not. Prepared waters used for testing included distilled water, a high (1000 ppm) and a low (45 ppm) calcium solution, and a 39 ppm sodium water. Uncompacted plugs were constructed by dropping bentonite tablets into waterfilled cylinders, or by mixing powdered bentonite with preselected water volumes in order to obtain controlled initial water contents. The hydraulic conductivity of all plugs tested with all waters would result in a classification of practically impervious, by conventional soil mechanics standards. Variations of several orders of magnitude of the hydraulic conductivity are observed. Clay plugs constructed from bentonite tablets hydrated with unfiltered Ogallala groundwater exhibited reduced swelling and lower hydraulic conductivity than similar plugs constructed from tablets and distilled water. The differences in observed conductivities may be governed by factors such as swelling characteristics and permeant colloidal matter clogging plug pore spaces. Constant pressure injection and transient pulse testing methods have been tried to determine hydraulic conductivity. Especially in constant pressure injection tests, outflow volumes may require adjustment to account for consolidation drainage. Attempts made here at such corrections have not been successful. Consolidation testing requires extremely long time periods to approach a constant limit. It appears probable that the flow tests have not been pursued for a sufficient time to assure complete determination of water chemistry effects on hydraulic conductivity.

15a KEY WORDS AND DOCUMENT ANALYSIS

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